Atmospheric and Environmental Research, Inc.

# MODELING DEPOSITION OF ATMOSPHERIC MERCURY IN WISCONSIN

Prepared for

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# EXECUTIVE SUMMARY

This report summarizes a modeling sensitivity study, designed to gauge how estimates of mercury deposition in Wisconsin and neighboring states respond to changes in source emissions from those states. The research was sponsored by the Wisconsin Utilities Association (WUA), managed by the Electric Power Research Institute (EPRI), and conducted by Atmospheric and Environmental Research, Inc. (AER). The work draws on the experience of leading research scientists with expertise in air quality, mercury source inventories, and atmospheric chemistry and modeling.

The mercury inventory used was developed by EPRI for the entire United States, but modified for Wisconsin alone to include source terms from a State of Wisconsin mercury emissions inventory. EPRI estimates of utility mercury emissions, however, were used throughout for geographic consistency and to provide an equivalent basis for the utility source scenario terms.

The atmospheric mercury modeling system used incorporated models at three spatial scales: a global scale for initial conditions, with 8º x 10º grid cells; a continental-scale dispersion model, with 100 km grid spacing; and a sub-continental simulation at 20 km spacing. Model performance was evaluated by comparing wet deposition of mercury calculated by the model with annual deposition observations of the Mercury Deposition Network (MDN) stations nationally and for Wisconsin, for the same grid cells.

- The primary model used in this study (TEAM) simulated the transport, chemical and physical transformations, and deposition of mercury emissions using detailed chemical, meteorological, precipitation and geographic data.

- The model simulations focused on the upper midwest and the northeastern U.S. The model was initialized to incorporate the most recent inventory of mercury sources by the Wisconsin Department of Natural Resources, as well as regional meteorological and geographic data, and estimates of mercury contributions mapped from national inventories and global source estimates.

- In addition to a baseline case incorporating the full national and Wisconsin inventory, three emissions scenarios were modeled:
    - ◊ All Wisconsin mercury emissions set to zero.
    - ◊ All Wisconsin mercury emissions plus all coal utility boiler emissions from Minnesota, Iowa, Illinois, Indiana, Michigan, Missouri, and Ohio set to zero.
    - ◊ All Wisconsin coal utility boiler emissions set to zero.

- The study mapped the estimated inventory of both point sources (e.g., coal-fired power plants) and areas sources (e.g., motor vehicles) within Wisconsin. For other states, motor vehicle emissions were not explicitly simulated. Emissions are estimated to be highest in the southeastern part of Wisconsin, near the cities of Madison and Milwaukee. This is driven by the impact of area sources of mercury, which were distributed according to population density.

- The model compared simulations of wet mercury deposition to actual measurements obtained from 30 Mercury Deposition Network (MDN) stations (four of which are located in Wisconsin). Although the model overpredicts deposition at the Wisconsin sites by an average of 22%, this constitutes a reasonable measure of model performance for this level of spatial resolution.

- The modeling concluded that 4-10% of mercury deposited at the four Wisconsin MDN sites comes from the combined total of Wisconsin sources, and that 6-18% comes from Wisconsin sources plus regional power plant emissions.

- The modeling estimates that when Wisconsin utility emissions are completely eliminated, wet mercury deposition declines by less than 4% at the Wisconsin MDN sites and by less than 5% over most areas of the state.

- None of these estimates have been adjusted for model uncertainty, or systematic overprediction. Model uncertainty is believed to be due to inexact mercury emission inventories (and their unknown mercury chemistries) and to uncertain power plant plume chemical reactions. In the latter case, both laboratory and field measurements suggest that ionic forms of mercury in power plant plumes undergo reactions that rapidly convert these forms into the elemental form, which, in turn, does not significantly deposit locally or regionally. Thus, the mercury deposition simulations conducted in this study are likely to represent an upper bound on the contribution of local and regional sources to mercury deposition in Wisconsin.

- As part of this study, the results of two mercury modeling studies using EPA mercury models were reviewed and compared to the TEAM findings. The first study was conducted by U.S. EPA in the mid-1990's, and used the RELMAP model. The second study was funded by the Lake Michigan Air Directors Consortium (LADCO) and used the REMSAD model. The EPA study was part of the comprehensive Mercury Study Report to Congress, required as part of the 1990 Clean Air Act Amendments. The LADCO study was released in January of this year. The TEAM simulation performed here showed better performance when compared to MDN mercury wet deposition data than both the RELMAP and REMSAD modeling studies.

- The modeling results correspond to a snap-shot in time, but so far represent the most extensive study completed, to date, that begins to inform policy-makers of the complexity of attributing mercury in the environment to specific emission sources. The results confirm the regional, national, and even global scope of the mercury issue.

# 1. INTRODUCTION

The objective of this study is to estimate the effect of various emission sources on the atmospheric deposition of mercury (Hg) in Wisconsin. Any study with such an objective must first simulate the global cycling of Hg as well as its deposition on a finer continental/regional scale. This is necessary since Hg has an atmospheric lifetime on the order of one year (Schroeder and Munthe, 1998). The global simulation of Hg provides the boundary (i.e., upwind) condition for modeling Hg at continental or regional scales. In previous studies, EPRI conducted multi-scale simulations of the atmospheric fate and transport of Hg using a nested system of three modeling domains: a global model, a continental model over North America, and a regional model encompassing Wisconsin, its neighboring states, and other states in the northeastern United States (EPRI, 2000, 2002). This system is termed "one-way": results at a given model scale drive initial and boundary conditions at the next-smaller nested scale, but smaller scales do not determine larger-scale results. These models and their applications have been described by Seigneur *et al.* (2001a, 2002). Overall, the model performance was judged to be satisfactory for calculated ambient concentrations of elemental Hg [Hg(0)], divalent gaseous Hg [Hg(II)], and particulate Hg [Hg(p)], as well as for wet deposition fluxes of total Hg. In particular, use of the nested grid system improved model performance for locations in Wisconsin with deposition data for comparison (Seigneur *et al.*, 2002).

In the study described here, the multi-scale modeling system described in EPRI (2002) and Seigneur *et al.* (2002) was used to conduct a one-way nested grid simulation for the year 1998. The coarse grid simulation results from an earlier EPRI study (EPRI, 2002) were used to provide boundary conditions for the inner fine grid. The fine grid used in this study has a horizontal resolution of 20 km and covers the upper midwestern and northeastern United States. The base case emission inventory for the state of Wisconsin used in the above-mentioned EPRI study was modified (except for utility coal-fired boiler emissions) to reflect the emission inventory data available from the Wisconsin Department of Natural Resources (WDNR, 2001). The WDNR emission inventory dates to the 1994-95 period. The emission inventory developed for all U.S.

power plants by EPRI (2001) was utilized for the above referenced EPRI study (EPRI, 2002) as well as for the present study.

Three emission sensitivity simulations were also conducted using the fine grid domain. In the first sensitivity simulation, all anthropogenic Hg emissions from Wisconsin were set to zero. In the second sensitivity simulation, all anthropogenic Hg emissions from Wisconsin and all coal-fired power plant Hg emissions from Minnesota, Iowa, Illinois, Indiana, Michigan, Missouri, and Ohio were set to zero. In the third sensitivity simulation, all coal-fired power plant Hg emissions from Wisconsin were set to zero.

When interpreting the results of these simulations it is important to recognize that TEAM is a regional model designed to study impacts at larger scales, approximately the size of U.S. states. This model is not designed to simulate localized impacts of point sources at the grid-cell level as precisely as are atmospheric dispersion models, which more realistically represent plume behavior due to wind and temperature. Consequently, such local impacts are likely to be misrepresented by the regional model, as discussed in Section 2.2. Section 2 provides a brief description of the model including the modeling domain, the modified emissions inventory and other model inputs. Simulation results are presented in Section 3. This section includes a description of the spatial distribution of deposition values and a comparison of model results with observations for the base case. The sensitivity of the simulated mercury deposition values to the three different emission scenarios is also discussed. The results of this study are compared with those obtained in studies conducted for the Lake Michigan Air Directors Consortium (LADCO), a consortium comprised of state air regulatory agencies of the upper Midwest, and for the U.S. Environmental Protection Agency (EPA). In these two studies, Hg deposition simulations were conducted with the EPA-supported REMSAD and RELMAP models, respectively. The results of these studies along with important considerations for study interpretation are also summarized in Section 3, and the simulation results and interpretations are discussed in Section 4.

# 2. MODEL DESCRIPTION

## 2.1 Modeling Domain and Grids

Figure 2-1 shows the location of the nested fine grid (regional domain) with respect to the outer grid (continental domain). The outer grid is based on a polar stereographic projection with a horizontal resolution of about 100 km in each direction (EPRI, 2000; Seigneur *et al*., 2001a). The continental domain includes northern Mexico, the contiguous United States, and southern Canada, and consists of 47 grid rows and 63 grid columns. This modeling domain serves as a source of background concentrations for the inner fine grid via the boundary.

The inner grid, also based on a polar stereographic projection, has a horizontal resolution of about 20 km in each direction and consists of 80 grid rows and 120 grid columns (EPRI, 2002; Seigneur *et al*., 2002). Figure 2-2 shows the inner grid domain in more detail. This domain encompasses Wisconsin (an area of 140,000 $km^2$, which corresponds to about 400 model grid cells), its neighboring states, other states in the northeastern United States, and parts of the provinces of Ontario and Quebec in Canada. Six layers with different resolution are used in the vertical direction from the surface to about 6 km. The vertical resolution is finer in the layers closer to the surface than in the higher layers – four of the six layers are within 2 km of the surface.

## 2.2 Air Quality Model

The model used in this study is the <u>T</u>race <u>E</u>lements <u>A</u>nalysis and <u>M</u>odeling (TEAM) system. It has been described in detail previously (Pai *et al*., 1997; Seigneur *et al*., 2001a, 2002); therefore, only a brief summary is presented here. TEAM is a 3-D Eulerian model that simulates the transport, chemical and physical transformations, wet deposition and dry deposition of Hg species. The model simulates three Hg species: gaseous elemental Hg [Hg(0)], gaseous divalent Hg [Hg(II)], and particulate divalent Hg [Hg(p)]. The atmospheric transformations of these species are simulated with a mechanism that represents the current state-of-the-science. This mechanism, described

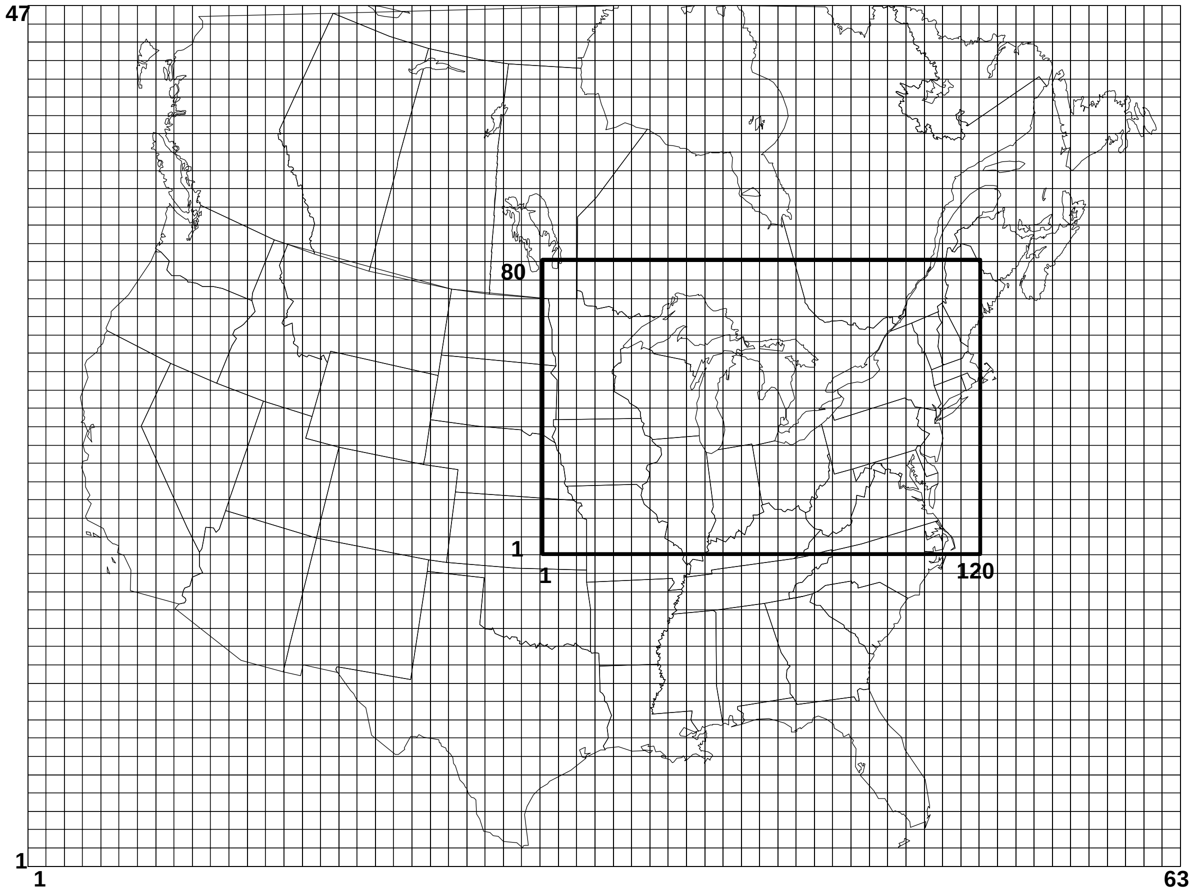



Figure 2-1. The nested grid system with the continental and regional domains.

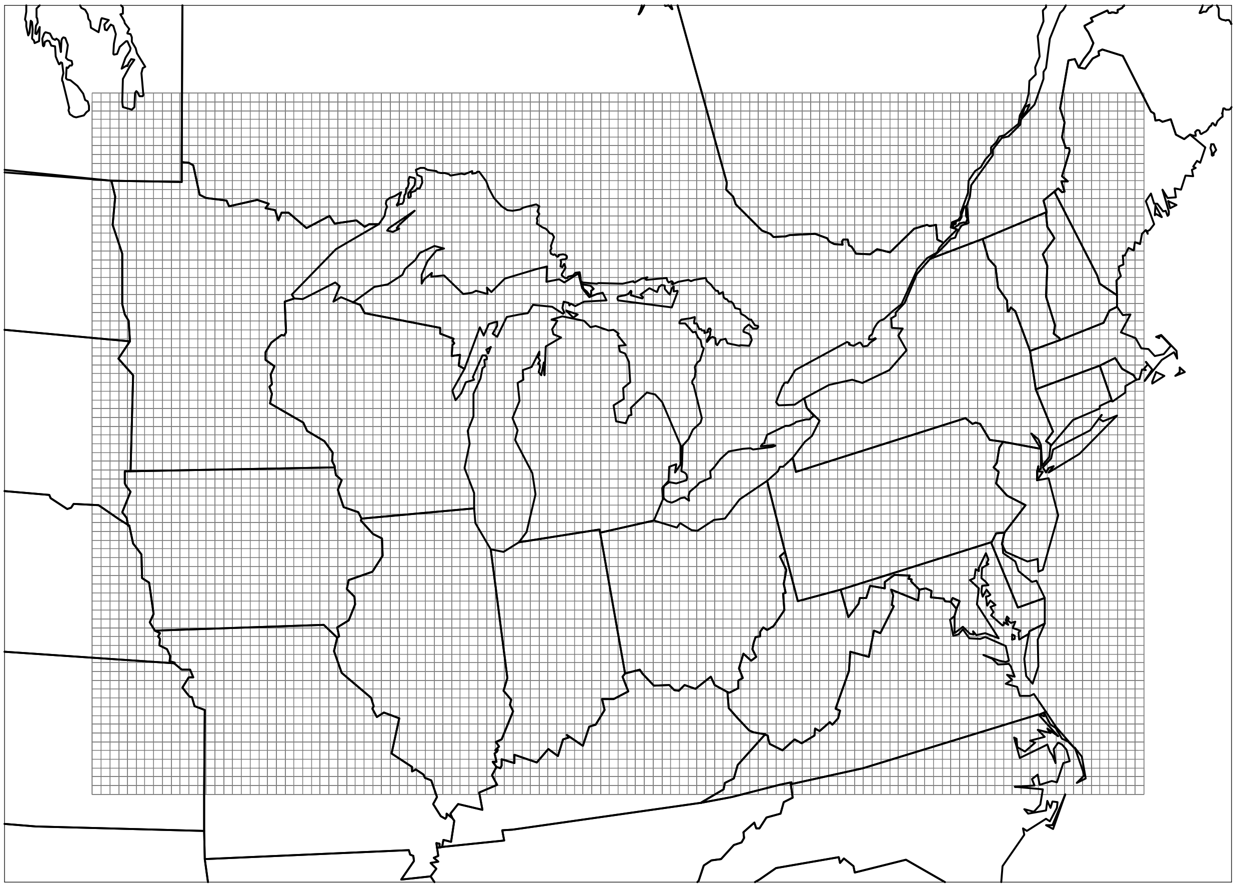


Figure 2-2. The regional (fine grid) domain.

by Seigneur *et al.* (2001a, 2002), includes gas-phase transformations, gas/droplet equilibria, ionic equilibria, solution/particle adsorption equilibrium, and aqueous-phase transformations of Hg. Of the three Hg species, Hg(0) is the least soluble and is not removed rapidly from the atmosphere; its atmospheric lifetime is currently estimated to be about one year. Hg(II) species are very soluble and removed rapidly from the atmosphere via dry and wet deposition processes; its atmospheric lifetime is currently estimated to be one to several days in the absence of clouds and on the order of hours or less in the presence of precipitating clouds. Hg(p) is present mainly in fine ($PM_{2.5}$) particles; as a result, it is not removed as fast as Hg(II) but its atmospheric lifetime is considerably shorter than that of Hg(0).

Transport processes in TEAM include movement of the Hg species by the 3-D mean wind flow (advection) and dispersion by atmospheric turbulence. Dry deposition is simulated using a resistance-transfer approach and, therefore, depends on underlying land use and meteorology in addition to the species being deposited. Wet deposition includes removal by both rainout (in-cloud scavenging) and washout (below-cloud scavenging) transient processes. Wet deposition is simulated only for Hg(II) and Hg(p) since Hg(0) is not very soluble (only about one one-millionth of Hg(0) is dissolved in cloud droplets and most Hg(0) remains in the gas phase).

It should be noted that TEAM is a regional model that is designed to study regional potential impacts of a variety of sources. The large volumes of the model grid cells (24 $km^3$ in the surface layer; greater aloft) lead initially to some artificial mixing of the plumes from point sources throughout the individual cells, since the joint source emissions in a single grid cell are instantaneously mixed within the grid cell volume. This treatment tends to rapidly mix the point source emissions to the ground, whereas in reality the point source plume (including Hg) is likely to stay elevated above ground level for significant distances downwind (e.g., Karamchandani et al., 2002). TEAM is likely to overestimate the local ground-level impacts of such sources. Therefore, the results of this modeling study should not be seen as representative of the local impacts of individual sources. Other models, such as the Reactive and Optics Model of Emissions (ROME) (Constantinou et al., 1995) or the Total Risk of Utility Emissions model (TRUE) (Constantinou and Seigneur, 1993), should be used to assess local impacts.

## 2.3 Emissions Inventory

The emissions estimates used in this study are from a hybrid inventory. The domain-wide inventory is based on a scenario for 1998 developed by AER (Seigneur *et al*., 2001a; EPRI, 2002) for EPRI, with a new inventory inserted for the state of Wisconsin. This new Wisconsin inventory is based on the inventory for 1994/1995 provided by WDNR (2001). The previous AER emission inventory contained approximately 2.1 Mg/y of Hg emissions for the state of Wisconsin. The new inventory has total Hg emissions of 3.0 Mg/y, including many area sources not present in the AER inventory.

Table 2-1 provides a breakdown of the two emission inventories according to source category. All emissions for Wisconsin, except for those due to coal-fired power plants, are from the state inventory. As previously stated, the emissions due to coal-fired power plants were obtained from EPRI values and were used in the original inventory and in this hybrid inventory to provide a consistent basis for evaluating the effects of zeroing out the emissions of coal-fired power plants outside of Wisconsin. It should be noted that the EPRI inventory value for Hg emissions from coal-fired power plants in Wisconsin differs by less than 1% from the WDNR inventory value.

In the hybrid inventory used for this study, the largest sources are coal-fired power plants (940 kg/yr), chloralkali production (505 kg/yr), medical waste incineration (273 kg/yr), and combustion of petroleum products (233 kg/yr). When the new emission rates from the WDNR inventory are used, the total Hg emissions for the domain bounded by the fine grid change from 81.3 to 82.2 Mg/y.

The emissions estimates for many of the source categories are quite different between the two inventories. For example, the AER inventory does not have emissions due to chloralkali production whereas it is a large source of emissions for the new Wisconsin inventory. This difference is because the lone chloralkali facility in Wisconsin did not report emissions to the EPA's Toxic Release Inventory (TRI) for 1998. Therefore, it was not included in the AER inventory. Other differences, such as the larger emission rates for non-utility coal burning and medical waste incineration in the AER inventory are likely due to different emission factors being used in the preparation of the two inventories. It is important to note that with the exception of the coal-fired

Table 2-1. Emission Inventories by Source Category (kg/yr) in Wisconsin.

| **Source Type** | **AER / EPRI Inventory** | **WDNR Inventory** |
|---|---|---|
| Coal-fired Power Plants | 940 | 940[a] |
| Other Coal Combustion | 417 | 190 |
| Non-utility Petroleum Product Combustion | 32 | 233 |
| Wood Burning | 7 | 5 |
| Municipal Waste Incineration | 78 | 80 |
| Medical Waste Incineration | 439 | 273 |
| Pulp & Paper | 52 | 2 |
| Lamp Breakage | 95 | 49 |
| Lime Manufacturing | 3 | 58 |
| Human Cremation | 9 | 17 |
| Chloralkali | 0 | 505 |
| Mobile Sources | 0 | 105 |
| Other Point Sources | 0 | 155 |
| Other Area Sources | 0 | 373 |
| **Total** | **2072** | **2985** |

(a) AER / EPRI inventory value, Wisconsin DNR value was 947 kg/yr.

power plant source category, we have little, if any, speciated mercury measurements for Wisconsin source categories. The lack of speciated mercury measurements for these other source categories increases the uncertainty associated with modeled mercury deposition.

Figure 2-3 provides a map of the new emissions inventory for Wisconsin. Figures 2-4 and 2-5 break down these total emissions into those derived from point and area sources, respectively. Point sources were assigned to the model grid cells according to their latitude/longitude coordinates. For area sources, the following methodology was used. For source categories that were already present in the AER inventory, the emissions were scaled to reflect the WDNR value and the spatial distribution remained unchanged. For source categories that were not present in the AER inventory, the spatial distribution of those area sources was assumed to follow the population spatial distribution. This is driven by the impact of area sources of Hg. Point source emissions are dispersed around the state more evenly, but area sources, many of which are distributed according to population, tend to be clustered toward the southeastern corner of the state, near the cities of Madison and Milwaukee. Emissions for both point and area sources are lowest in the northern portion of the state. The result is that overall emissions are highest in southeastern Wisconsin.

## 2.4 Other Model Inputs

Other model inputs for the regional domain shown in Figure 2-2 for the year 1998 have been described in detail by Seigneur *et al.* (2002) and, hence, are discussed briefly below. The 3-D wind and temperature fields were obtained from the results of a simulation of the Nested Grid Model (NGM), a prognostic meteorological model applied by the National Oceanic and Atmospheric Administration (NOAA). Cloud and precipitation fields were obtained from the National Center for Atmospheric Research (NCAR). Annual precipitation data were obtained for 1998 from the National Acid Deposition Program (NADP)/Mercury Deposition Network (MDN) for stations in the U.S. while precipitation data for Canada were obtained from the Canadian Climate Network. The concentrations of the chemical species reacting with Hg, namely, $O_3$, OH,

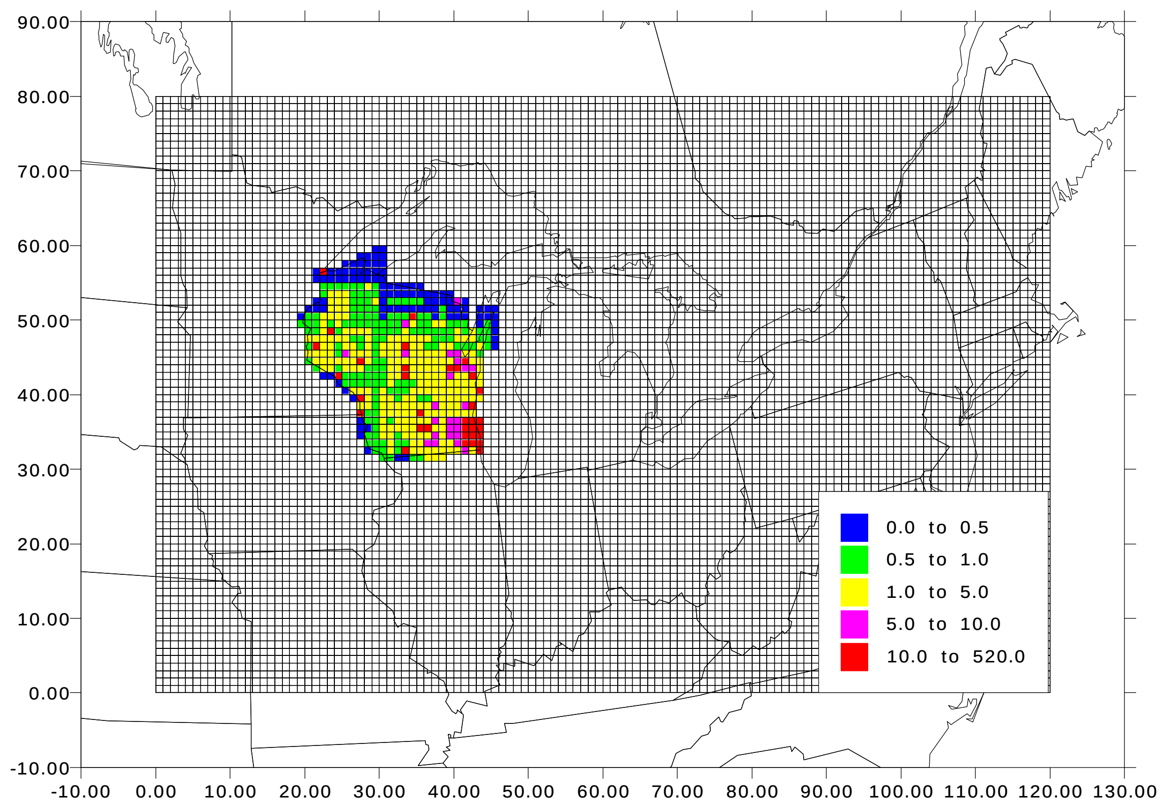



Figure 2-3. Total annual emissions of mercury (kg/yr) in Wisconsin.

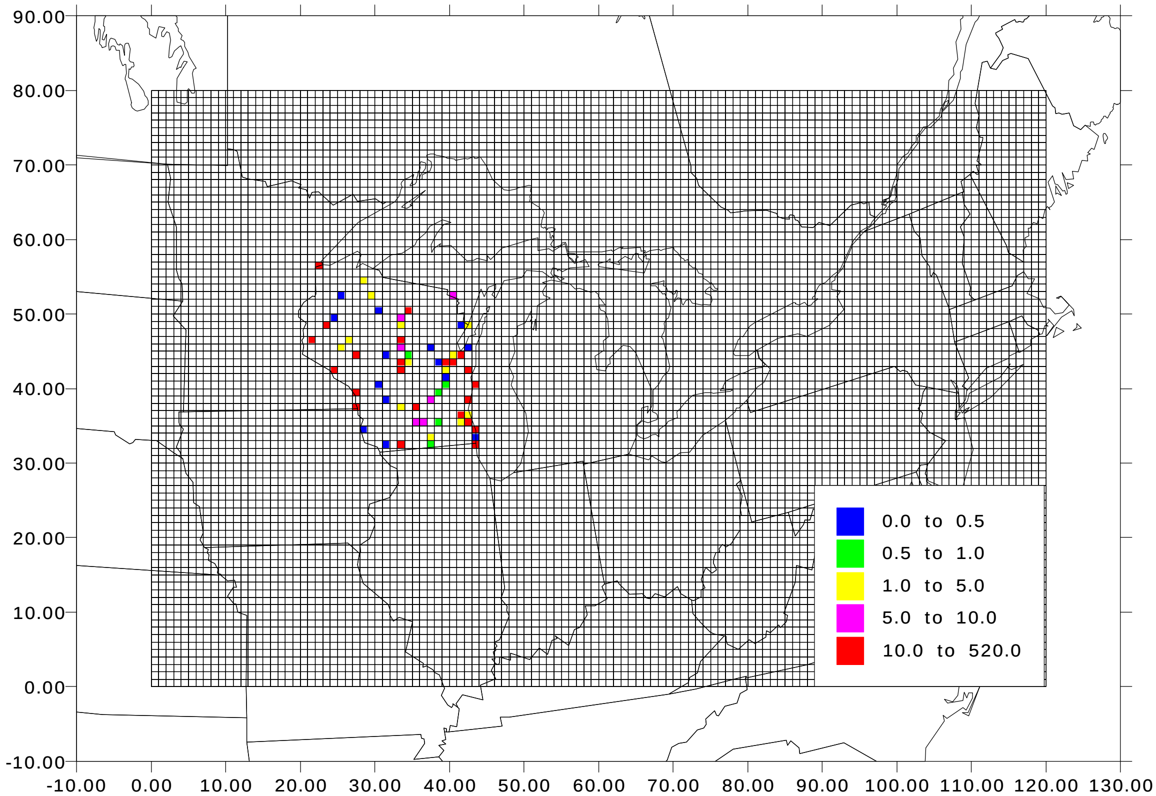



Figure 2-4. Point source emissions of mercury (kg/yr) in Wisconsin.

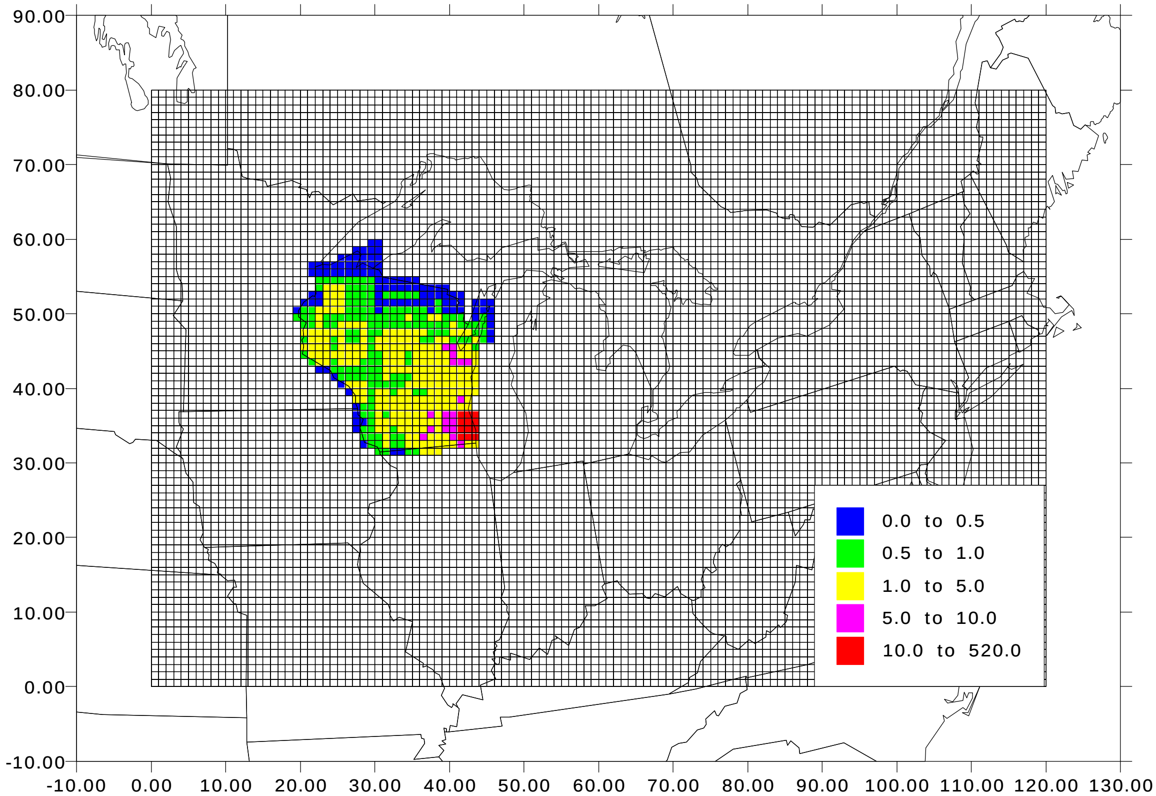



Figure 2-5. Area source emissions of mercury (kg/yr) in Wisconsin.

$HO_2$, $H_2O_2$ and $SO_2$ were obtained from the same 3-D fields as those used for the global Hg simulation described in Seigneur *et al.* (2001a). The concentrations of $Cl_2$, HCl and atmospheric PM in droplets used in this study are the same as those listed in Seigneur *et al.* (2001a).

The boundary conditions consist of the concentrations of Hg(0), Hg(II), and Hg(p) as a function of location, height, and season. Boundary conditions for the fine grid simulations were developed using the results from the coarse grid simulations of Seigneur *et al.* (2001a, 2002). The hourly concentrations at each coarse grid cell along the boundaries of the inner grid were extracted from the 3-D concentration fields of the coarse grid simulation to create spatially varying time-dependent boundary condition files for the inner grid.

# 3. SIMULATION RESULTS

## 3.1 Base Case

A base-case TEAM simulation was conducted over the fine grid domain for the 1998 calendar year using the inputs described in the previous section. The spatial distribution of the dry, wet, and total deposition values of Hg simulated in this base case are discussed below. We only considered deposition of Hg(II) and Hg(p) since Hg(0) deposition is typically not taken as an input to watershed/lake models (e.g., Harris et al., 1996).

### 3.1.1 Spatial distribution of Hg deposition

The simulated Hg dry deposition flux over the modeling domain is illustrated in Figure 3-1. Dry deposition fluxes are less than 5 $\mu g/m^2$-yr over most of Wisconsin. Values range between 5 and 10 $\mu g/m^2$-yr near Madison, areas surrounding Milwaukee, along the Minnesota border, and near Green Bay. The highest dry deposition flux in Wisconsin (15-30 $\mu g/m^2$-yr) occurs near Milwaukee and in the western part of the state. Dry deposition patterns typically tend to follow the spatial distribution of emission sources of Hg, including area sources, which are primarily a function of population patterns. This is evident from the Wisconsin emission tile plots shown in Figures 2-3 through 2-5. Local sources tend to have a larger impact on dry deposition because this process depends directly on the atmospheric concentration of Hg. While this is a reasonable generalization, local deposition is also highly dependent on the magnitude of these source emissions and the relative fractions of Hg(II), Hg(p), and Hg(0), which vary among source categories. Dry deposition fluxes are less than 5 $\mu g/m^2$-yr in most of Minnesota, Iowa, and Missouri (except highly urbanized areas such as Minneapolis-St. Paul and St. Louis). The regions with the highest dry deposition fluxes over the domain are in the Northeast, e.g., the Ohio Valley, Pennsylvania, West Virginia, Maryland, New Jersey, Connecticut, Massachusetts, and the southern part of New York State. Urban areas such as Chicago and Detroit also show high dry deposition.

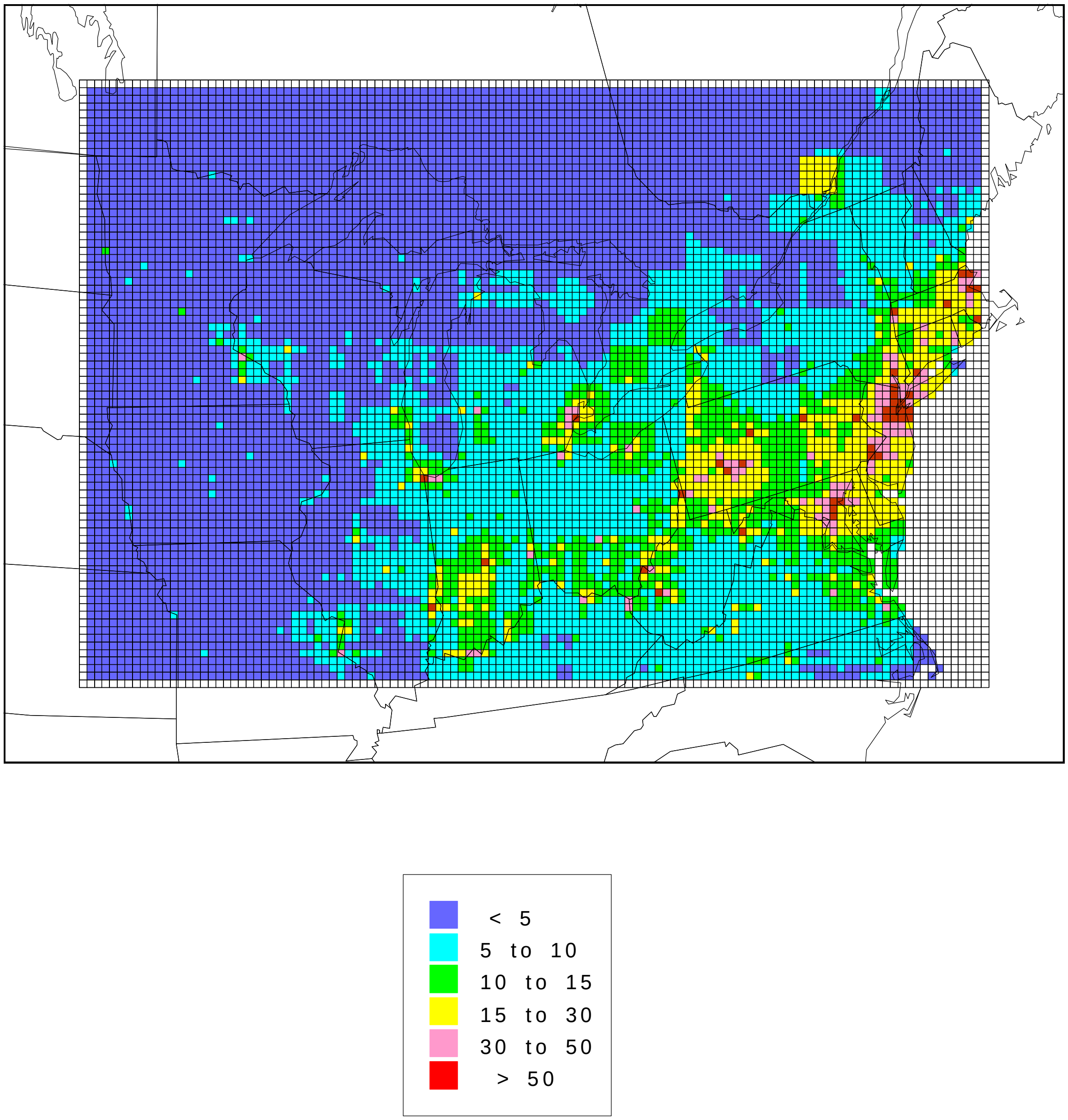


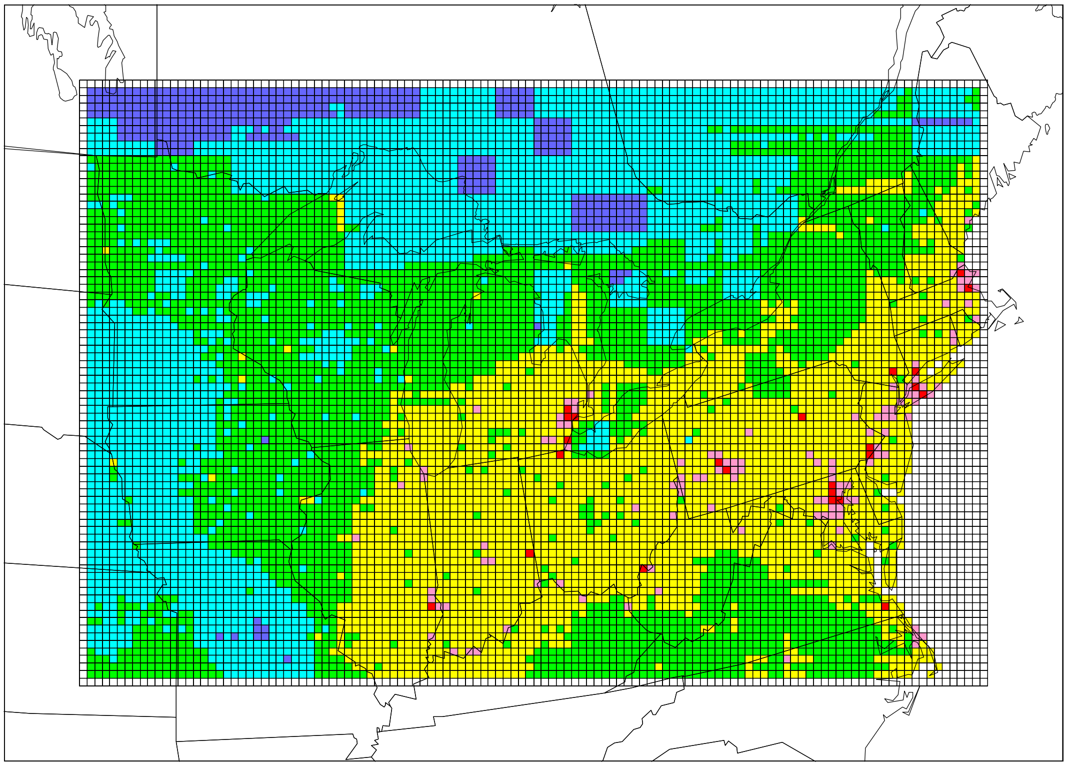

Figure 3-1. Simulated dry deposition flux of Hg ($\mu g/m^2$-yr) over the modeling domain.

Figure 3-2 presents the simulated Hg wet deposition flux over the modeling domain. Wet deposition fluxes in most of Wisconsin are in the range of 10 to 15 $\mu g/m^2$-yr. However, wet deposition exceeds 15 $\mu g/m^2$-yr near Green Bay and in areas near Milwaukee and along the southeastern border with Illinois. It is likely that there is some contribution from Hg transported from the northern area of Illinois. Unlike dry deposition, wet deposition depends on the occurrence of a precipitation event, which may not occur until Hg has traveled some distance. Wet deposition fluxes are between 5 and 15 $\mu g/m^2$-yr in most of Minnesota, Iowa, and Missouri. Similar values are also obtained for most of Canada, upstate New York, southern Virginia, and northern Michigan. Wet deposition fluxes above 15 $\mu g/m^2$-yr are simulated over most of Indiana, Ohio, Pennsylvania, Maryland, New Jersey, and New England with isolated areas of very high wet deposition.

The simulated total Hg deposition flux (i.e., wet and dry combined) is depicted in Figure 3-3. Total Hg deposition fluxes are in the range of 10 to 20 $\mu g/m^2$-yr in most of Wisconsin. The fluxes exceed 20 $\mu g/m^2$-yr in a few areas near Milwaukee and along the Minnesota border. The latter is likely due to emission sources near Minneapolis-St. Paul, Minnesota. Values are less than 20 $\mu g/m^2$-yr in most of Minnesota, Iowa, and Missouri. Total Hg deposition fluxes in Canada are less than 20 $\mu g/m^2$-yr except in southern Ontario and near Montreal where they can exceed 30 $\mu g/m^2$-yr. Hg deposition fluxes exceed 20 $\mu g/m^2$-yr over most of the Northeast with large fractions of Pennsylvania, West Virginia, Maryland, New Jersey, and New England exceeding 30 $\mu g/m^2$-yr. Some grid cells along the Ohio Valley, in Pennsylvania, West Virginia, Maryland, New York, Massachusetts, Connecticut, and Indiana show deposition fluxes exceeding 100 $\mu g/m^2$-yr.

### 3.1.2 Model performance

The simulated Hg wet deposition fluxes for 1998 were compared with measurements at all the sites of the Mercury Deposition Network (MDN, 2000) for which data were available for 1998. The 1998 MDN database includes 27 sites in the United States (including four in Wisconsin) and 3 sites in Canada. The MDN network of



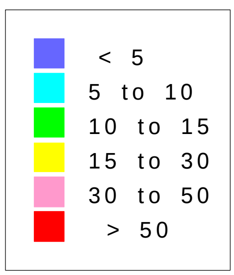



Figure 3-2. Simulated wet deposition flux of Hg ($\mu g/m^2$-yr) over the modeling domain.

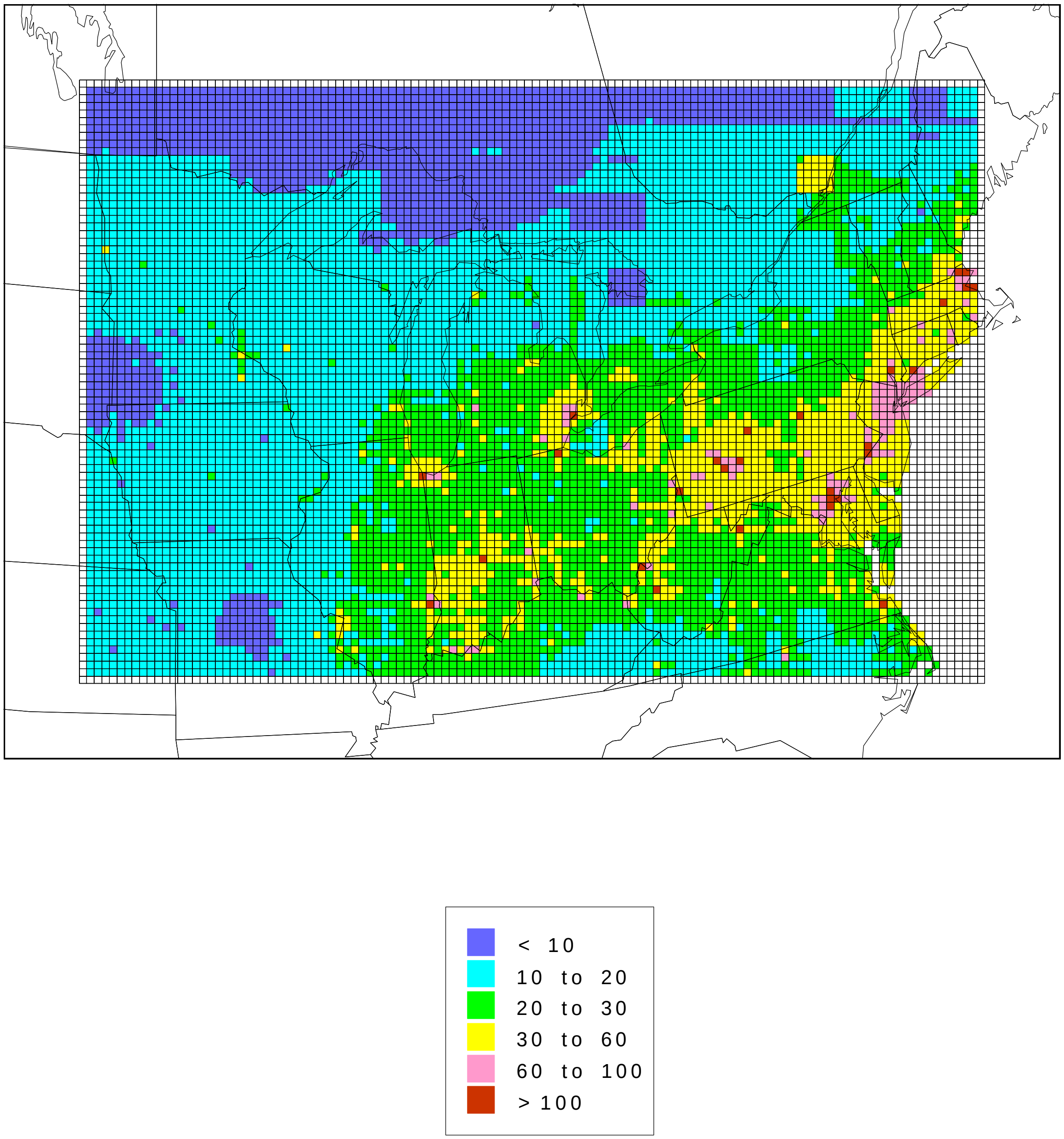



Figure 3-3. Simulated total deposition flux of Hg (µg/m$^2$-yr) over the modeling domain.

stations is illustrated in Figure 3-4. As part of model performance evaluation, fine grid simulation results were used for MDN sites within the fine grid domain, while for stations outside this domain, the previous coarse grid simulation results of Seigneur *et al.* (2001a) were used.

The coefficient of determination ($r^2$) is 0.12, the normalized bias is 22%, and the normalized gross error is 44% when deposition fluxes are compared by state or province (Figure 3-5) (normalized error = $\frac{1}{N}\sum_{i=1}^{N}\left|\frac{P_i - O_i}{O_i}\right|$; normalized bias = $\frac{1}{N}\sum_{i=1}^{N}\left(\frac{P_i - O_i}{O_i}\right)$; $P_i$ = prediction, $O_i$ = observation; $N$: number of samples). The relatively poor correlation is due primarily to the large overestimates of deposition occurring in Pennsylvania and New Hampshire. In this comparison, when more than one MDN station was located in a state, the average value of the wet deposition data across stations was used for that state. Variability in the MDN values within a state ranged from 13% for Pennsylvania (two stations) to 44% for Wisconsin (four stations). Other factors contributing to the over-prediction of Hg have been discussed by Seigneur *et al.* (2002); the implications for the present study will be discussed later.

When wet deposition fluxes are compared at specific sites (Figure 3-6), the coefficient of determination is 0.10, the average bias is 25%, and the gross error is 45%. The black dots used as symbols in Figure 3-6 indicate the four Wisconsin sites. The model predictions at these Wisconsin sites are presented in Table 3-1. The model tends to slightly over-predict wet deposition at these monitoring stations. The normalized error ranges from 4% to 36%. Overall, the normalized gross bias (and error) is 22% at the four Wisconsin sites.

This simulation is based on the precipitation patterns for one calendar year, in this case, 1998. Both MDN annual measurements of wet deposition and model simulations will vary from year to year as a result of differences in precipitation amounts and patterns. In essence, this simulation represents one "snap-shot in time".

The results obtained with the AER/EPRI emission inventory were presented in EPRI (2002). Those results showed lower values of simulated wet deposition at the Wisconsin grid points that are in closer agreement with the MDN wet deposition

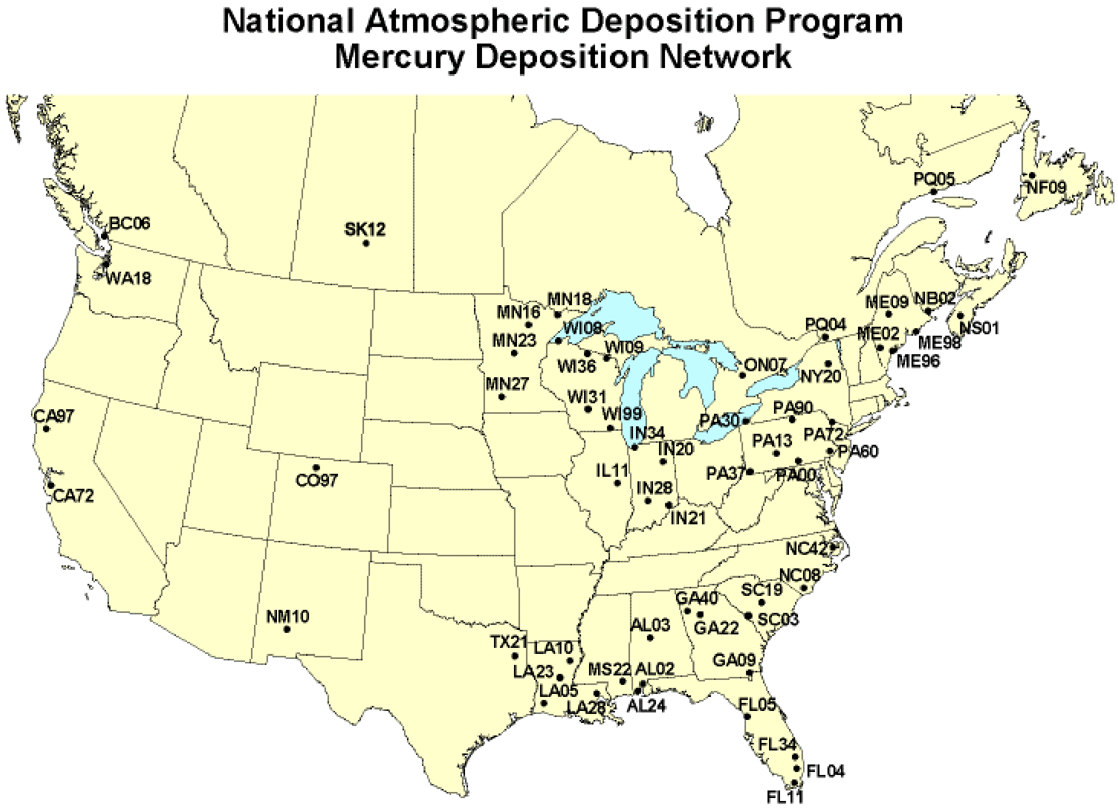



Figure 3-4. Map of Hg deposition monitoring stations in the MDN network (MDN, 2000) (Note: Sites in Wisconsin operational in 1998 were WI08, WI09, WI36, WI99).

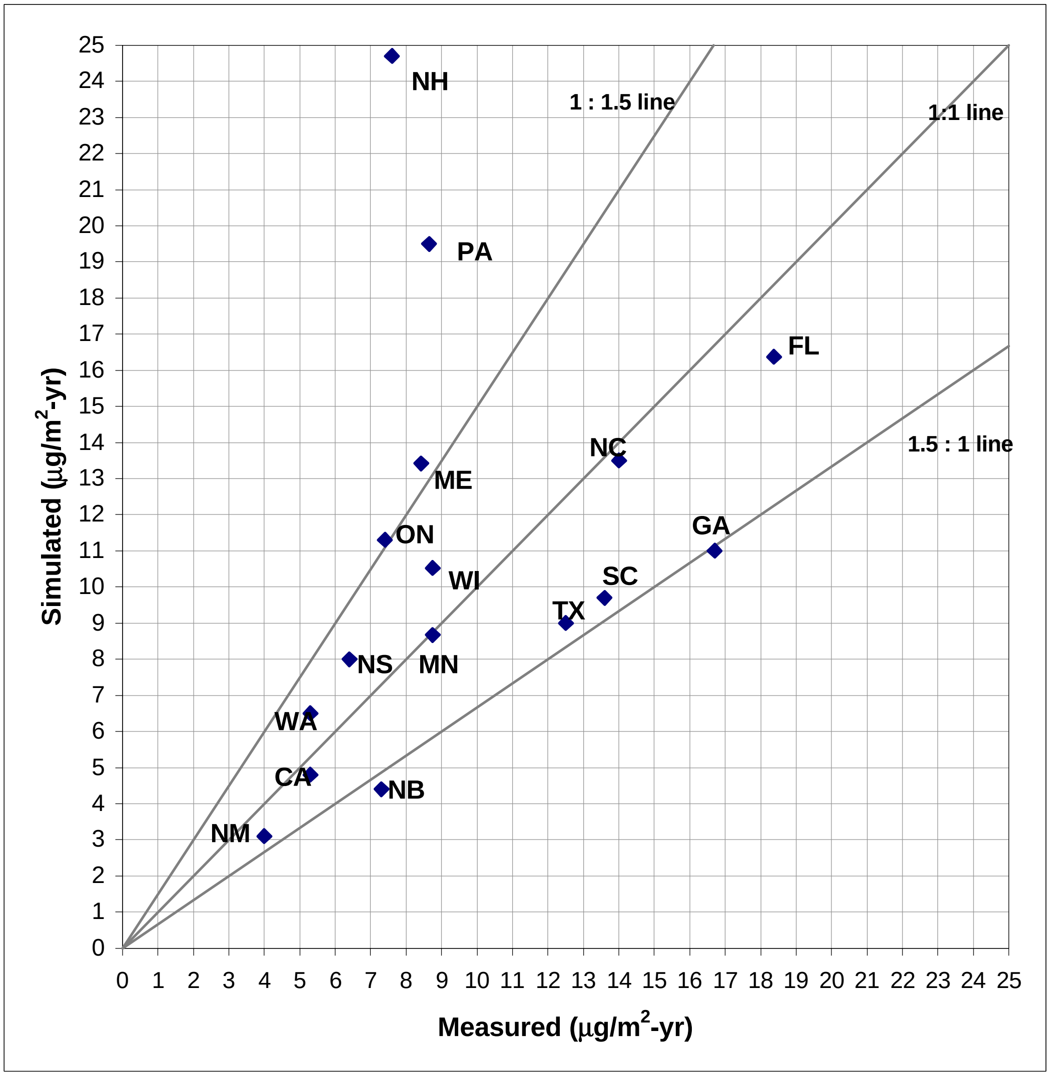



Figure 3-5. Comparison of measured and simulated Hg wet deposition fluxes (µg/m$^2$-yr) in 1998 by state and province.

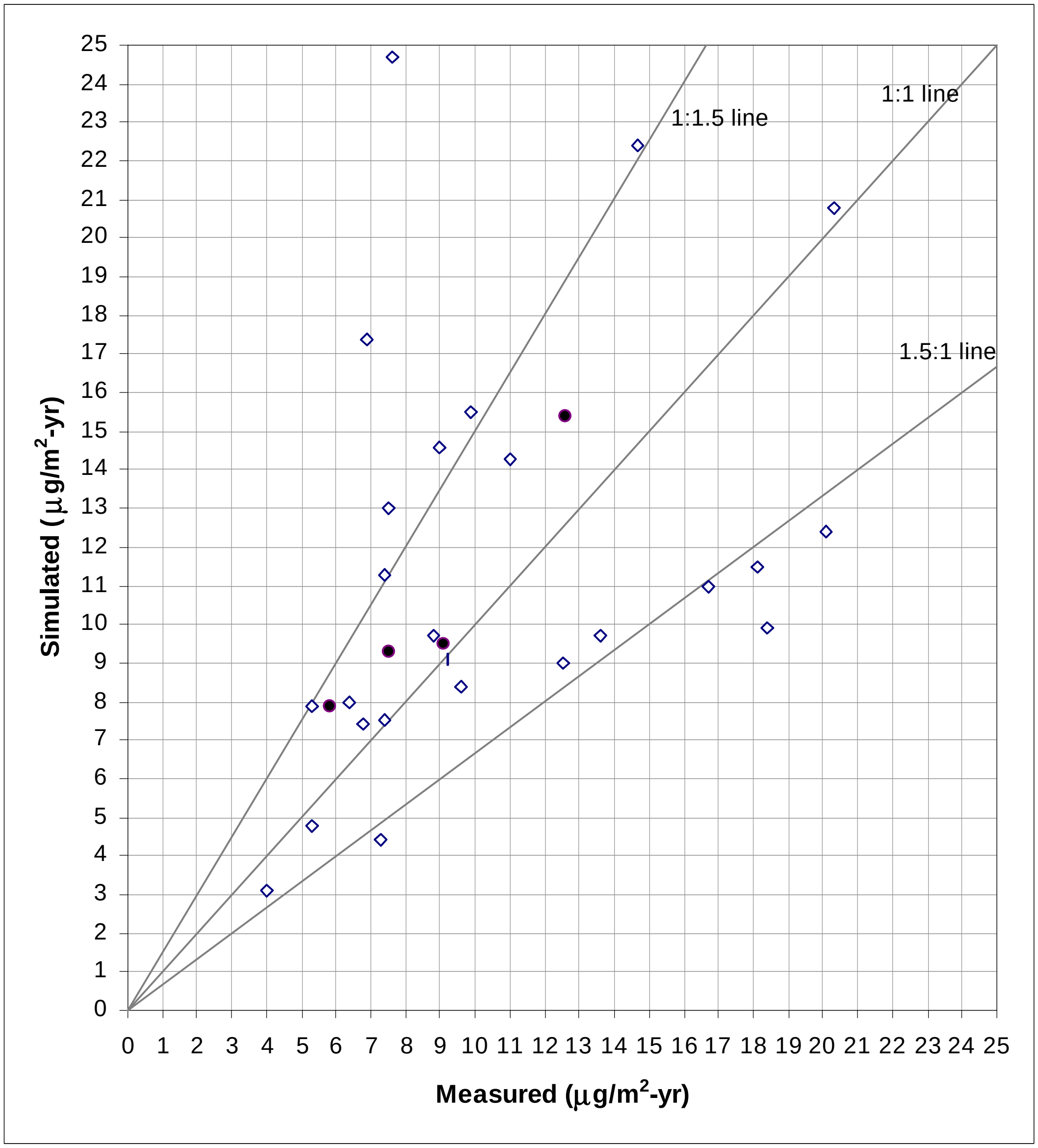



Figure 3-6. Comparison of measured and simulated Hg wet deposition fluxes (μg/$m^2$-yr) in 1998 at MDN sites (black dots indicate sites in Wisconsin).

Table 3-1. Comparison of measured and simulated Hg wet deposition fluxes in Wisconsin during 1998.

| MDN Site | Location | Measurement ($\mu g/m^2$-yr) | Simulation ($\mu g/m^2$-yr) | Normalized difference |
|---|---|---|---|---|
| WI08 | Brule River | 9.1 | 9.5 | 4% |
| WI09 | Popple River | 5.8 | 7.9 | 36% |
| WI36 | Trout Lake | 7.5 | 9.3 | 24% |
| WI99 | Lake Geneva | 12.6 | 15.4 | 22% |

measurements than the results for this study. Those lower values result from the lower Wisconsin emissions used in the EPRI (2002) study.

### 3.1.3 Comparison with REMSAD results

A memorandum from ICF Consulting to LADCO summarizes the application of the Regional Modeling System for Aerosols and Deposition (REMSAD) to the Midwest (ICF, 2002). Version 6.2 of REMSAD was used for this application. This section presents a comparison of the Hg wet deposition simulated by TEAM and REMSAD.

It should be noted that the REMSAD application discussed below differs from the TEAM simulation given earlier in several ways. The REMSAD study simulates the dry and wet deposition of Hg for the 1996 calendar year. The REMSAD coarse and fine grids have a horizontal resolution of about 36 km and 12 km respectively. The meteorology was driven by wind fields derived from an MM5 model simulation available from EPA. Hg emissions were obtained from EPA. Unlike TEAM, REMSAD pre-assigns values to the boundary concentrations of Hg based on the global background. Hg(0) was set to 1.7 $ng/m^3$ and Hg(II) and Hg(p) to much smaller values. REMSAD explicitly simulates the dry deposition of Hg(0) unlike TEAM which assumes Hg(0) background emissions and dry deposition to balance each other over North America. These differences should be kept in mind when reading the discussion that follows. We also understand that ICF Consulting is revising their simulation. The new results will likely change the comparison given below.

REMSAD underestimates the wet deposition of Hg at all MDN monitoring sites. The normalized bias is -77.4% and the normalized gross error is 77.4% at the 19 stations. Wet deposition fluxes are highly influenced by precipitation fields. TEAM uses daily precipitation fields from NCAR and refines them using annual precipitation data from the MDN database. REMSAD uses predicted precipitation fields from MM5 which may not be as accurate.

The REMSAD report also shows the simulated spatial distribution of wet and dry deposition of Hg. Simulated wet deposition fluxes of mercury are less than 2 $\mu g/m^2$-yr over most of Wisconsin. Regions with values between 2 and 4 $\mu g/m^2$-yr and small pockets with values greater than 5 μg/m$^2$-yr are present in the urban areas near Milwaukee, along the southeast border with Illinois, and along the border with Minnesota near St. Paul. While the general gradient of the wet deposition flux predicted by REMSAD in Wisconsin is similar to that of TEAM, the values simulated by the former are typically about 5 to 10 μg/m$^2$-yr lower than those simulated by TEAM.

The dry deposition fluxes of Hg simulated by REMSAD range from 5 to 10 μg/m$^2$-yr over most of Wisconsin. However, values in the northern part of the state are less than 5 μg/m$^2$-yr, which is consistent with the TEAM simulation. REMSAD simulates dry deposition fluxes that exceed 40 μg/m$^2$-yr all along the shores of Lake Michigan. This is due to the higher deposition velocities of Hg over water used by REMSAD. The highest dry deposition flux simulated by REMSAD exceeds 50 μg/m$^2$-yr in Wisconsin. On the other hand, the highest dry deposition flux simulated by TEAM in Wisconsin is in the range of 15 to 30 μg/m$^2$-yr at some isolated areas. In general the total deposition of Hg simulated by REMSAD is dominated by dry deposition, while in TEAM, the wet component has a slightly larger contribution than the dry component to Hg deposition in Wisconsin.

Finally, in its report to LADCO, ICF provides information on source allocation of certain Hg emissions. Specifically, the REMSAD simulation allocates between 1 and 5% of the Hg deposition occurring at the Wisconsin MDN sites to coal-fired power plants.

### 3.1.4 Comparison with RELMAP results

The U.S. Environmental Protection Agency (EPA) performed a study of the emissions, transport, and deposition of Hg in the U.S. based on their own emission inventory for 1994/5 and meteorological data for 1989 (EPA, 1997). The EPA study employed a model called the Regional Lagrangian Model of Air Pollution (RELMAP). The EPA also developed their own Hg emissions inventory for the 48 contiguous states. Their emissions inventory totaled 142 Mg/yr as compared to the AER/WI hybrid inventory total of 127 Mg/yr.

Because both the emissions inventory and meteorological data are for different years than those used in the AER study, it is difficult to draw any firm conclusions from a comparison of the results. However, a few trends do emerge. As shown in Table 3-2, the

Table 3-2. Comparison of estimates of Hg wet deposition ($\mu g/m^2$-yr) at MDN sites for TEAM and RELMAP simulations.

| **Station** | **MDN data (1998)** | **TEAM (1998)** | **MDN data[(a)] (1989)** | **RELMAP (1989/1994)** |
|---|---|---|---|---|
| MN16 | 8.8 | 9.7 | 5.94 | 3.37 |
| MN18 | 9.2 | 9.1 | 6.15 | 3.52 |
| NC42 | 9.9 | 15.5 | 19.41 | 11.52 |
| WI08 | 9.1 | 9.5 | 6.88 | 4.25 |
| WI09 | 5.8 | 7.9 | 7.91 | 4.48 |
| WI36 | 7.5 | 9.3 | 7.91 | 4.70 |

(a) The MDN data were adjusted to reflect modeled precipitation (EPA, 1997).

wet deposition of Hg in the Wisconsin area is higher in the TEAM simulations than in the RELMAP simulations. Dry deposition, on the other hand, compares quite favorably between the two simulations. The RELMAP simulations show dry deposition in Wisconsin to be between 1 and 10 $\mu g/m^2$-yr for most of the state and 10-30 $\mu g/m^2$-yr near Milwaukee. The TEAM simulations show the same trend.

The much larger differences in wet deposition than dry deposition between the two simulations may be due in part to different meteorological conditions in the two years simulated, but also to the use of an earlier mechanism of Hg chemistry in RELMAP.

The RELMAP simulation, which used fixed Hg speciation ratios for every U.S power plant, predicted that 6.7% of the Hg emitted from a large coal-fired power plant would deposit within 50 km of such a source (Table 5-15; in EPA, 1997).

Finally, the RELMAP study estimates that approximately 32% of the Hg(II) and 64% of the Hg(p) emitted in the United States is transported outside of the United States to become part of the global cycle.

### 3.1.5 Summary

The performance of TEAM appears to be significantly better than that of the other two models, REMSAD and RELMAP, particularly for the Wisconsin sites. When comparing model simulations and observations of Hg annual wet deposition, TEAM showed a normalized error of 44% whereas REMSAD showed a normalized error of 77%. When comparing model simulations and observations of Hg annual wet deposition at three Wisconsin sites, TEAM showed agreement with the observations within a factor of 1.4; RELMAP model simulations agreed with the observations within a factor of 1.8. Therefore, it appears warranted to use TEAM to estimate the regional impacts of Hg emission sources in Wisconsin and neighboring states.

## 3.2 Sensitivity Simulations

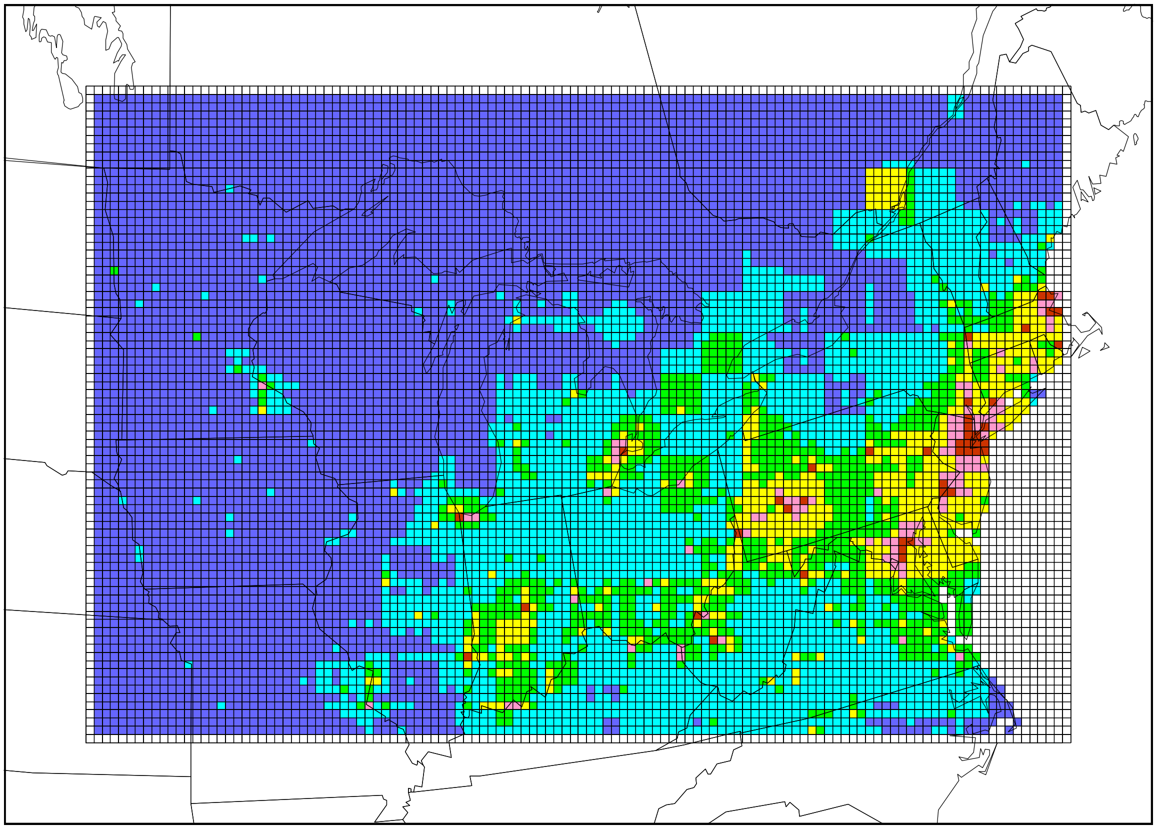
Three emission sensitivity simulations were conducted over the fine grid. In the first sensitivity simulation, all anthropogenic Hg emissions from Wisconsin were set to zero. In the second sensitivity simulation, all anthropogenic Hg emissions from Wisconsin and all coal-fired power plant Hg emissions from Minnesota, Iowa, Illinois, Indiana, Michigan, Missouri, and Ohio were set to zero. In the third sensitivity simulation, all coal-fired power plant Hg emissions from Wisconsin were set to zero. Table 3-3 presents a summary of the point and area source emissions for the base case and three emission sensitivity studies.

The dry, wet and total Hg deposition fluxes for the first sensitivity simulation (i.e., zero anthropogenic Wisconsin emissions) are presented in Figures 3-7, 3-8 and 3-9, respectively. The percent change in the simulated total deposition of Hg in the absence of anthropogenic Wisconsin emissions is illustrated in Figure 3-10. As seen from a comparison of Figures 3-7 and 3-1, simulated dry deposition drops to less than 5 $\mu g/m^2$-yr in all of Wisconsin except along the Minnesota border near St. Paul and on the shore of Lake Michigan on the Illinois border. Clearly Wisconsin experiences some dry deposition due to Hg emission sources in the Minneapolis-St. Paul area and areas to the north of Chicago. The impact of anthropogenic Wisconsin Hg emissions on wet deposition is seen from a comparison of Figures 3-8 and 3-2. Simulated wet deposition remains in the 10 to 15 $\mu g/m^2$-yr range in large parts of Wisconsin. Areas near Milwaukee drop from the 15-30 $\mu g/m^2$-yr range to the 10-15 $\mu g/m^2$-yr range due to zeroing out all anthropogenic Hg emissions in those areas.

The effect of all anthropogenic Wisconsin emissions on the total deposition flux of Hg is shown in Figures 3-9 and 3-10. As seen in Figure 3-10, most of the areas in Wisconsin exhibit a 10 to 25% decrease in total deposition with the following exceptions. The northern part of Wisconsin experiences less than 10% change. The impact on total Hg deposition exceeds 25% in areas around Milwaukee and Green Bay. Two isolated areas, near Milwaukee and in the western part of the state, show more than a 50% decrease. Typically the patterns of deposition decreases tend to follow the distribution of Wisconsin emissions seen in Figure 2-3. The higher reductions are driven primarily by the model simulating the elimination of the area source emissions, which occur in areas of the state having higher population density. These sources are represented in the Wisconsin state inventory as being directly proportional to population density.

Table 3-3. Summary of the point and area source emissions for the base case and three emission sensitivity studies.

| Scenario | Point source Hg emissions | | Area source Hg emissions | | Total Hg emissions | |
|---|---|---|---|---|---|---|
| | **(Mg/yr)** | **%reduction from base** | **(Mg/yr)** | **%reduction from base** | **(Mg/yr)** | **%reduction from base** |
| Base case | 66.5 | - | 15.7 | - | 82.2 | |
| No anthropogenic Hg emissions in Wisconsin | 64.4 | 3.2% | 14.8 | 5.7% | 79.2 | 3.6% |
| No anthropogenic Hg emissions in Wisconsin and no coal-fired power plant Hg emissions in nearby states | 53.1 | 20.2% | 14.8 | 5.7% | 67.9 | 17.4% |
| No coal-fired power plant Hg emission in Wisconsin | 65.5 | 1.5% | 15.7 | - | 81.2 | 1.2% |



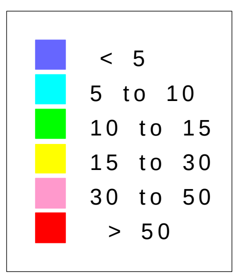



Figure 3-7. Simulated dry deposition flux of Hg ($\mu g/m^2$-yr) over the modeling domain with anthropogenic Hg emissions in Wisconsin set to zero.

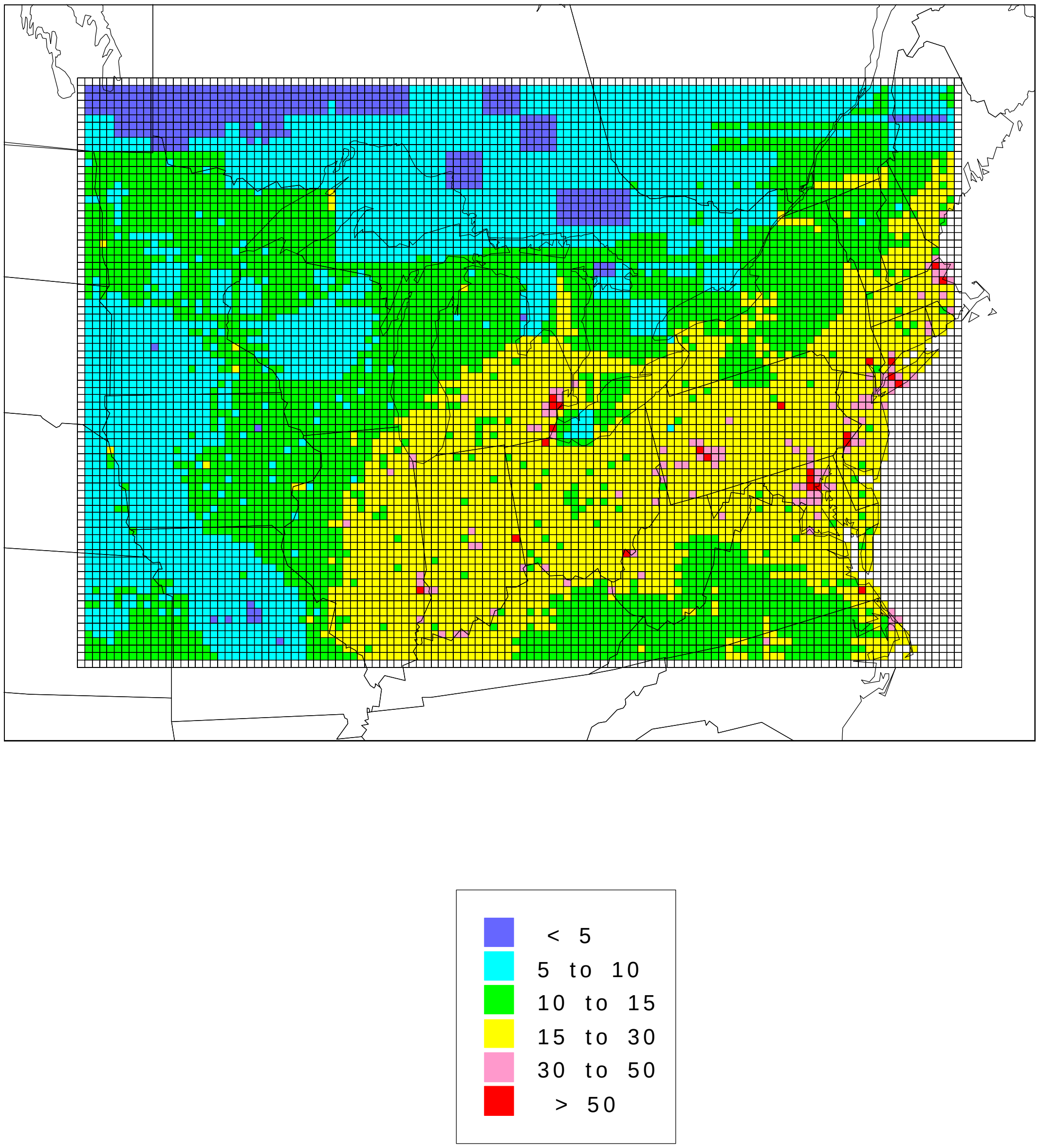
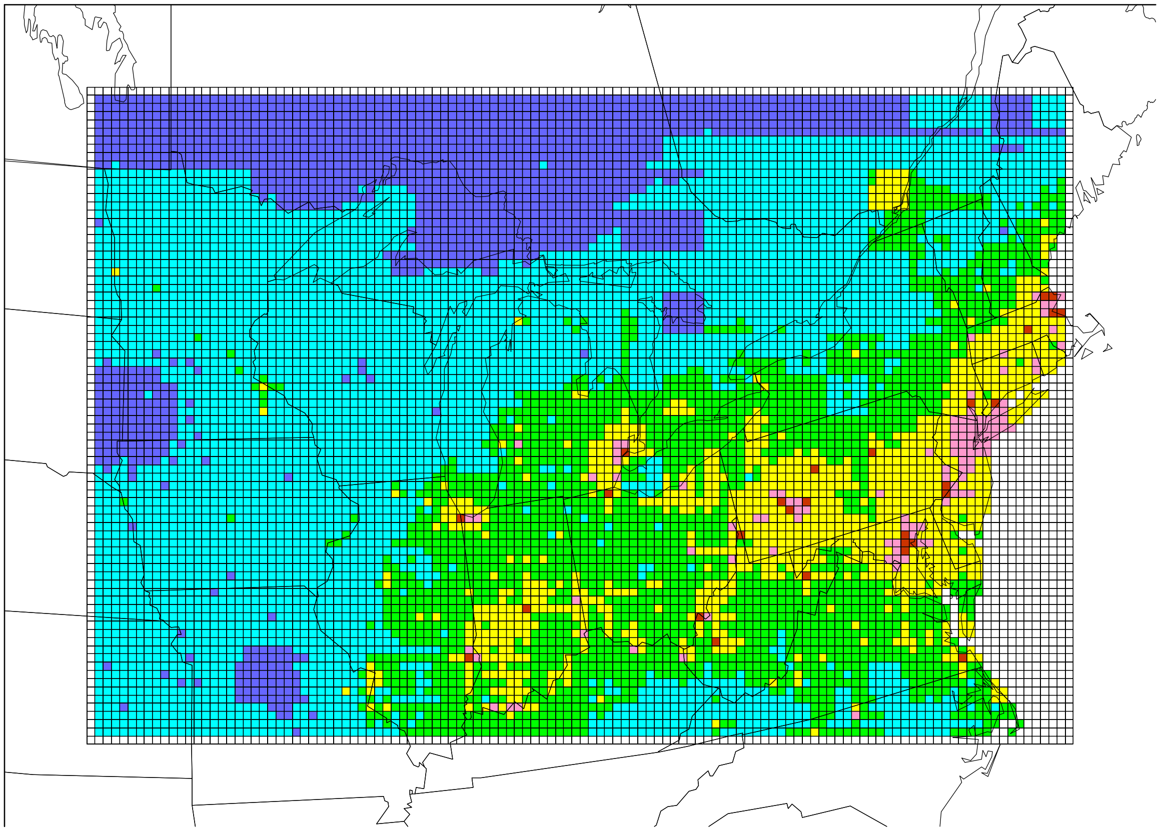



Figure 3-8. Simulated wet deposition flux of Hg (µg/m$^2$-yr) over the modeling domain with anthropogenic Hg emissions in Wisconsin set to zero.



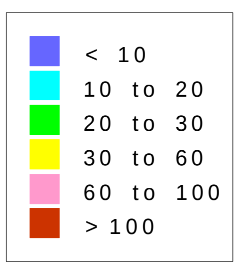



Figure 3-9. Simulated total deposition flux of Hg ($\mu g/m^2$-yr) over the modeling domain with anthropogenic Hg emissions in Wisconsin set to zero.

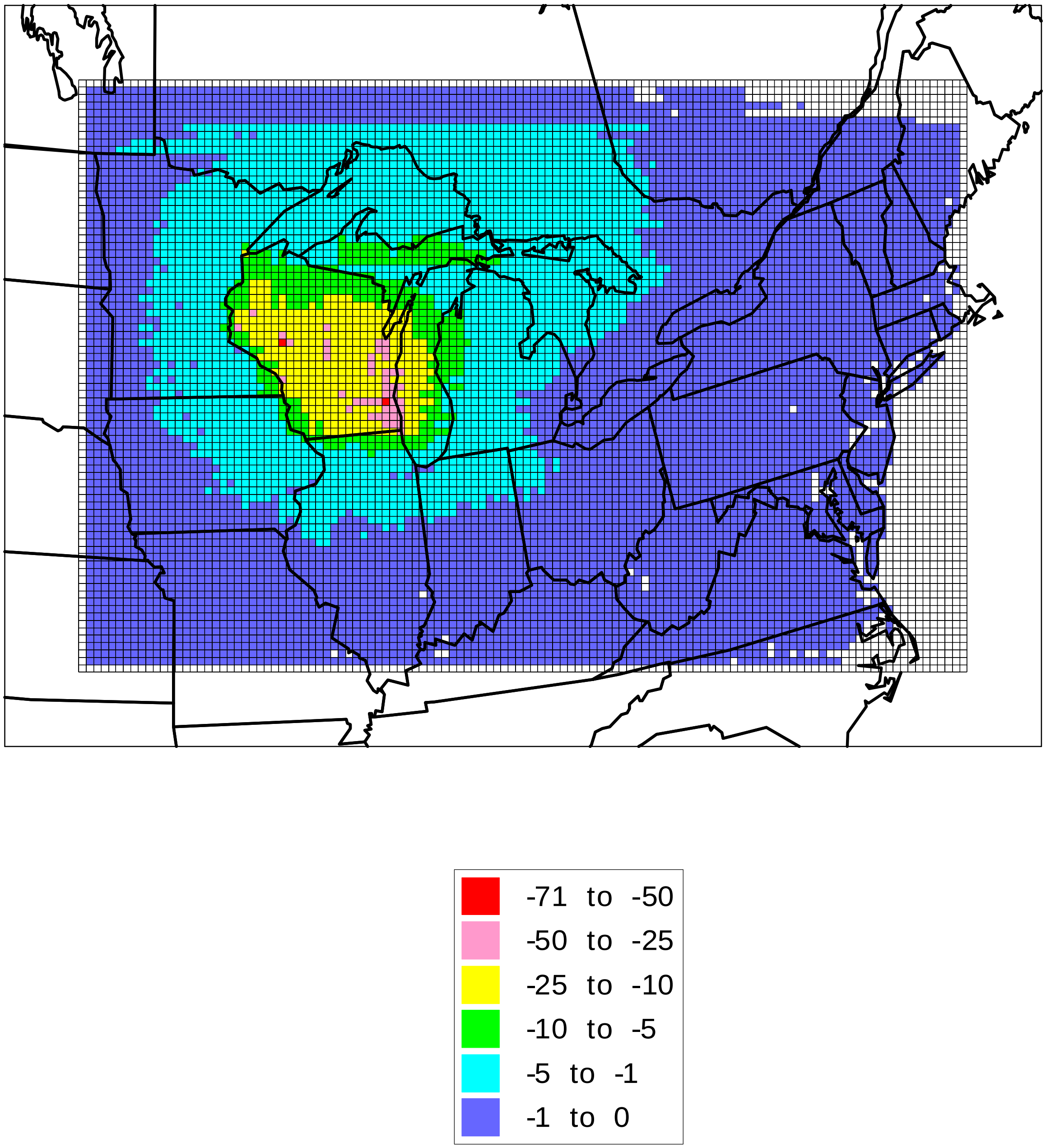


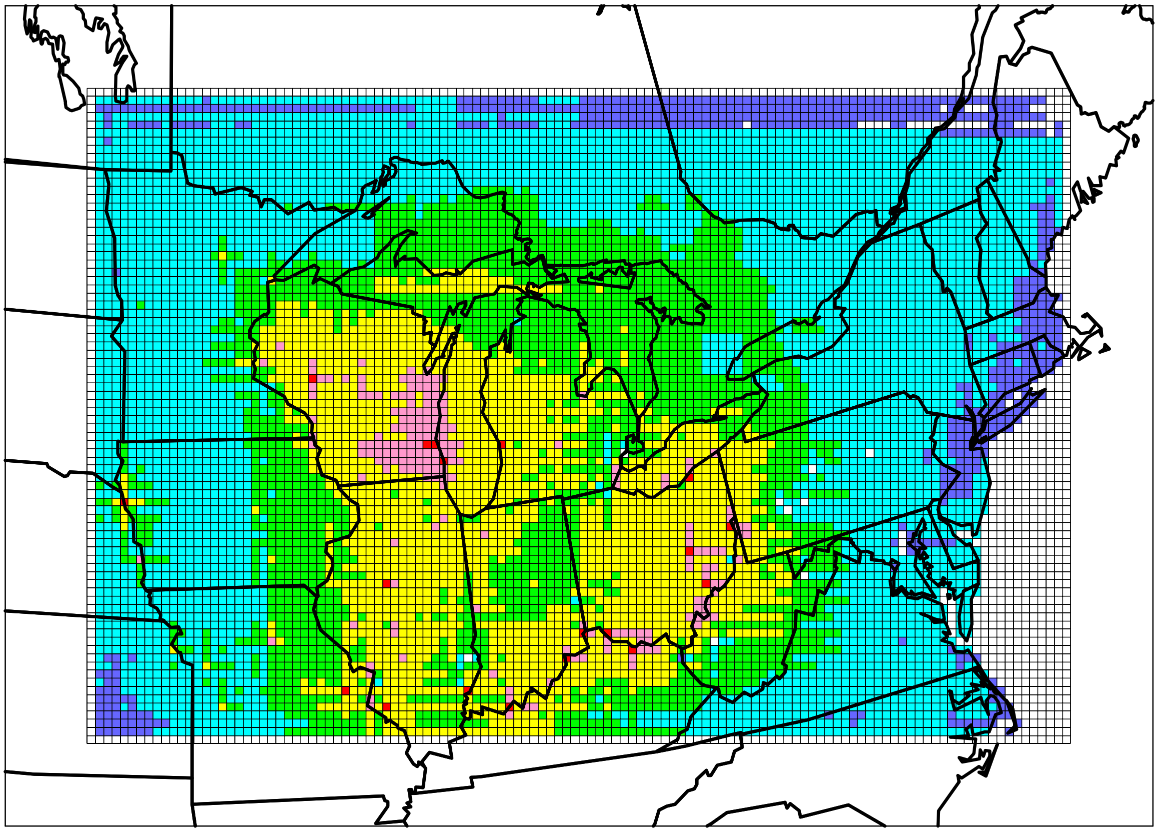

Figure 3-10. Percent change in the simulated total deposition flux of Hg over the modeling domain with anthropogenic Hg emissions in Wisconsin set to zero. (Note that these regional model simulations do not properly characterize local impacts of individual point sources.)

The percent change in total Hg deposition flux for the second sensitivity simulation (i.e., zero anthropogenic Wisconsin Hg emissions and zero coal-fired power plant Hg emissions from nearby states) is presented in Figure 3-11. It can be seen from a comparison of Figure 3-11 with Figure 3-10 that utility emissions outside Wisconsin have an impact on areas surrounding Milwaukee. The areas exhibiting 25 to 50% decreases in total Hg deposition are larger in the second sensitivity simulation than in the first. Figure 3-12 shows coal-fired power plant Hg emissions in the states of Minnesota, Iowa, Illinois, Indiana, Michigan, Missouri, and Ohio. It is possible that emissions from utilities in some of the neighboring states of Minnesota, Iowa, Michigan, and Illinois have an impact on Hg deposition in Wisconsin. This issue needs to be investigated further.

Figure 3-13 illustrates the results of the third sensitivity simulation, setting Wisconsin coal-fired power plant Hg emissions to zero. Most of Wisconsin shows less than 5% decrease in total Hg deposition when coal-fired utility Hg emissions in Wisconsin are set to zero. The northwestern part of the state has less than a 1% decrease in Hg deposition. As expected, the pattern of Hg deposition reductions across the state follows the spatial distribution of utility emissions as shown in Figure 3-14. The results of this modeling cannot be used to draw conclusions about the local impacts of individual sources, because of the simplified treatment of point source emissions.

The greater percentage reductions (10 to 25%) that appear in individual grid cells on the shore of Lake Michigan and in the central region of the state reflect to some extent the simplified treatment of point source emissions in regional models such as TEAM. As noted in Section 2.2, the results of such regional model simulations are likely to overestimate the local impacts of individual point sources.

Table 3-4 summarizes the impact of the three emission scenarios of emissions changes on wet deposition of Hg at the four MDN sites in Wisconsin. Emissions of Hg from Wisconsin contribute less than 10% at the four MDN sites in Wisconsin. Emissions of Hg from coal-fired power plants in states near Wisconsin and in Ohio and Missouri contribute less than 8% at the MDN sites in Wisconsin. Emissions of Hg from coal-fired power plants in Wisconsin contribute less than 1% at three of the MDN sites in Wisconsin and less than 4% at the fourth site.



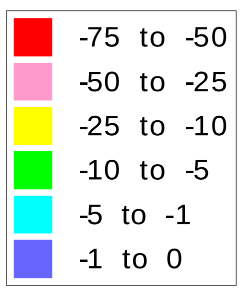



Figure 3-11. Percent change in the simulated total deposition flux of Hg over the modeling domain in the absence of anthropogenic Hg emissions in Wisconsin and coal-fired power plant Hg emissions in Minnesota, Iowa, Illinois, Indiana, Michigan, Missouri, and Ohio. (Note that these regional model simulations do not properly characterize local impacts of individual point sources.)

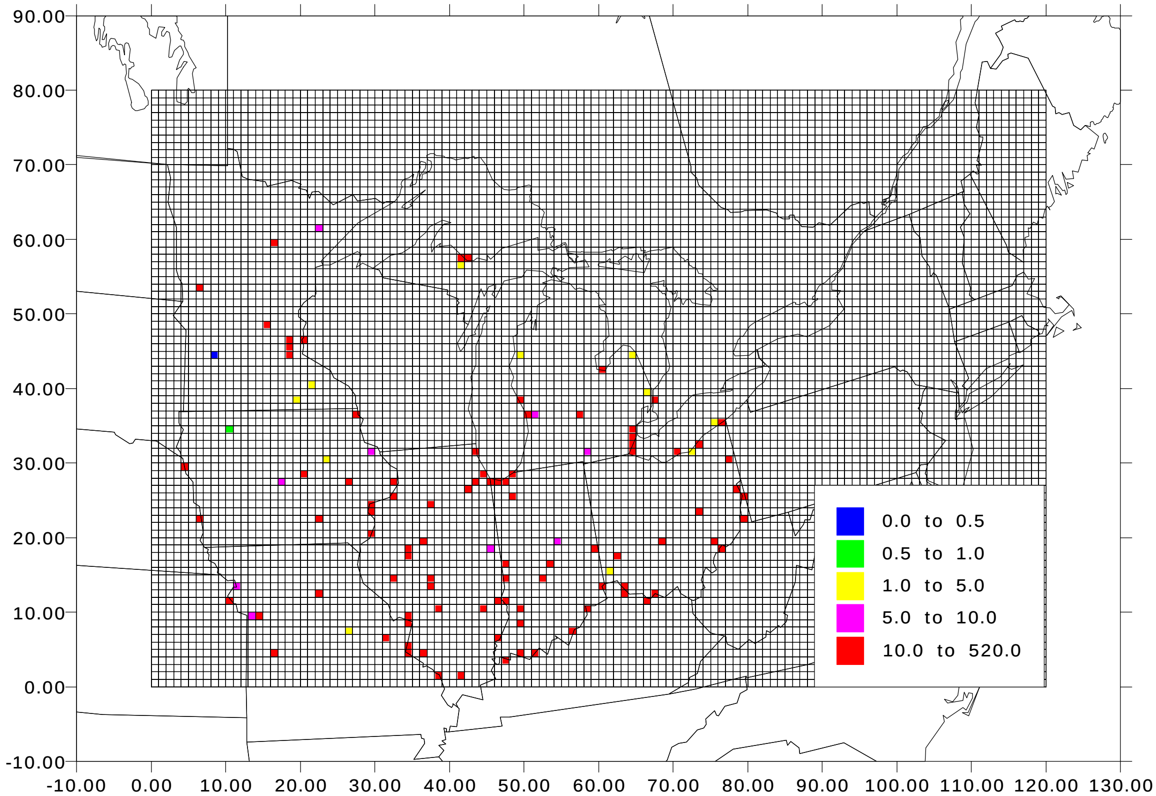



Figure 3-12. Spatial distribution of coal-fired power plant Hg emissions (kg/yr) in Minnesota, Iowa, Illinois, Indiana, Michigan, Missouri and Ohio.

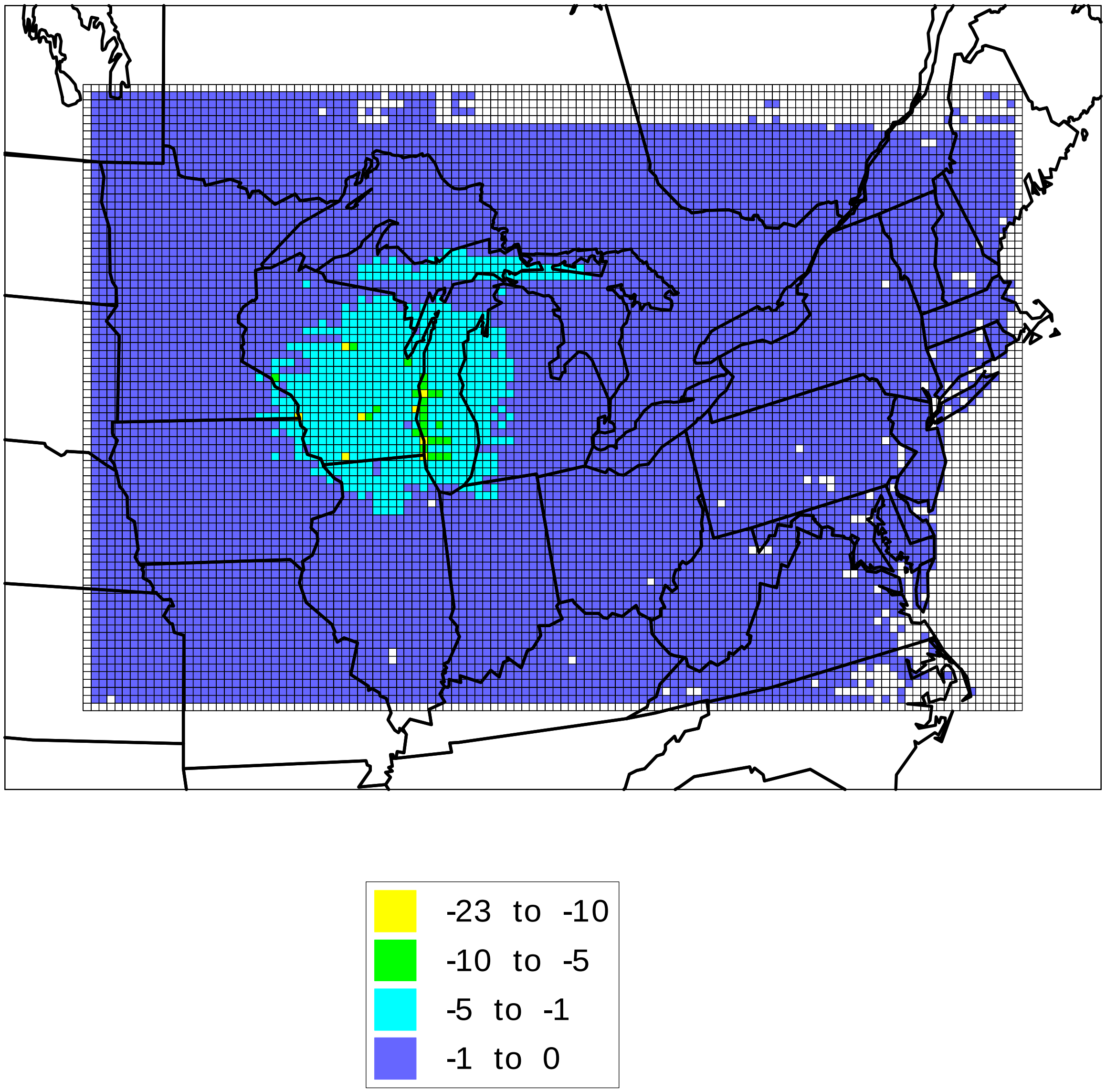



Figure 3-13. Percent change in the simulated total deposition flux of Hg over the modeling domain with coal-fired power plant Hg emissions in Wisconsin set to zero. (Note that these regional model simulations do not properly characterize local impacts of individual point sources.)

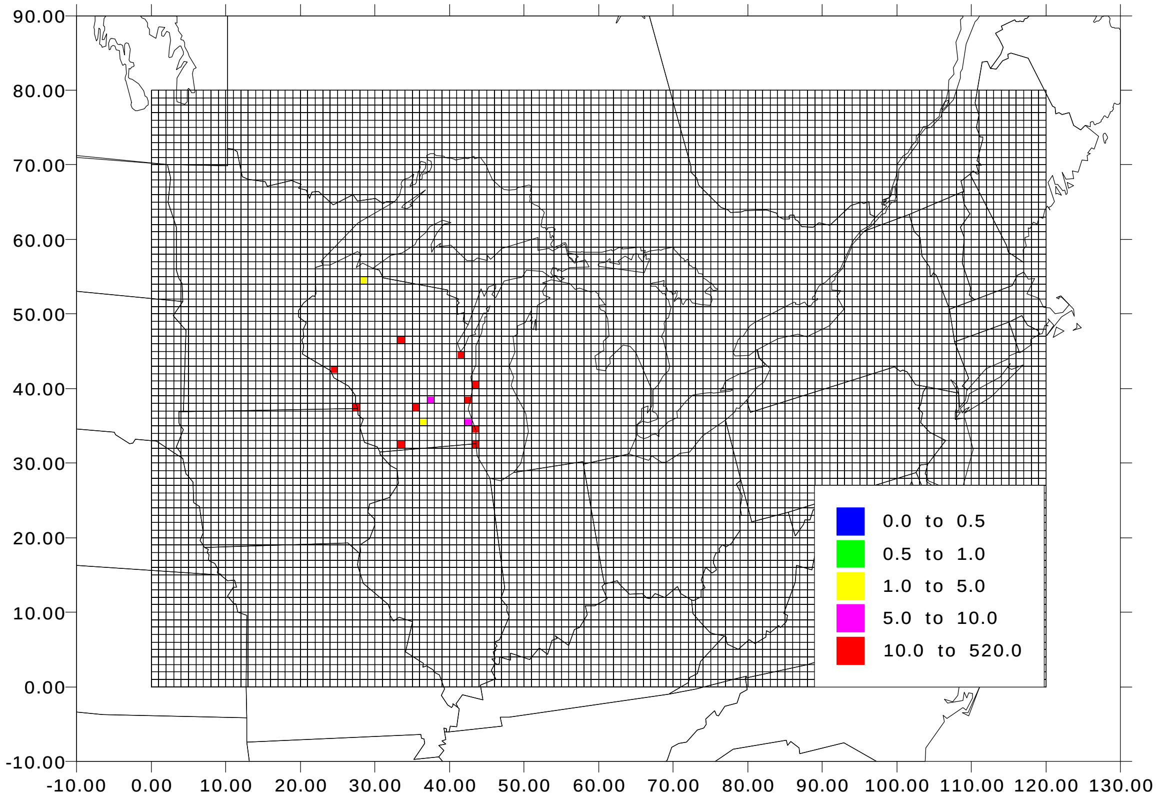



Figure 3-14. Spatial distribution of coal-fired power plant Hg emissions (kg/yr) in Wisconsin.

Table 3-4. Effect of local and regional emissions at Wisconsin MDN sites.

| MDN Site | No Wisconsin emissions | No Wisconsin and regional power plant emissions | No power plant emissions in Wisconsin |
|---|---|---|---|
| WI08 | -6% | -9% | -1% |
| WI09 | -5% | -8% | -1% |
| WI36 | -4% | -6% | -1% |
| WI99 | -10% | -18% | -4% |

These contributions are likely to be upper limits due to current uncertainties in Hg chemistry. These uncertainties are discussed below.

Seigneur *et al.* (2002) discussed the overprediction of Hg wet deposition in the northeastern U.S. and concluded that a possible source of uncertainty in the model is the treatment of the atmospheric transformations of Hg. The mechanism incorporated in TEAM represents our knowledge of Hg chemistry as of early 2001. It is likely that some reactions take place that are not currently in models and may have significant effects on the reduction and oxidation of Hg species. Specifically, chemical reduction of Hg(II) to Hg(0) by other substances emitted from power plants would lead to less deposition of Hg downwind of power plants, since Hg(II) deposition dominates total Hg deposition around Hg sources. As currently treated in regional Hg models, including TEAM, the reduction of $HgSO_3$ is too slow because of (1) the competing formation of $HgCl_2$ (Seigneur *et al.*, 1994) and (2) slow kinetics at low temperature (van Loon et al., 2000). The use of new equilibrium constants for aqueous-phase equilibria between $Hg^{2+}$, $SO_3^{2-}$ and $HgSO_3$ reported in the literature (van Loon *et al.*, 2001) could lead to higher $HgSO_3$ concentrations and, consequently, faster reduction of Hg(II) to Hg(0). This may explain, in part, the current overpredictions of Hg wet deposition in the northeastern United States.

There seems to be considerable corroborative evidence of reduction of Hg(II) to Hg(0) in power plant plumes from various experimental studies. First, the MDN data along a west-to-east transect from Minnesota to Pennsylvania show no significant spatial gradient in Hg annual wet deposition fluxes although the Ohio Valley includes several large Hg emission sources located, under prevailing wind conditions, upwind of Pennsylvania. Second, the University of North Dakota Energy and Environmental Research Center and Frontier Geosciences, Inc. conducted experiments where the exhaust flue gases from a coal-fired power plant stack were sampled, diluted and analyzed in a Teflon-lined dispersion chamber. These experiments showed a lower Hg(II)/Hg(0) ratio in the chamber than in the stack (Laudal, 2001). Third, ambient sampling of Hg species [Hg(II), Hg(0), and Hg(p)] downwind of coal-fired power plants in the Atlanta region suggests that the Hg(II)/Hg(0) ratio at the downwind location is lower than the Hg(II)/Hg(0) ratio estimated from the Information Collection Request (ICR) data for the stack emissions (Edgerton *et al.*, 2001). Furthermore, modeling of these power plant plumes indicates that current models do not account for this change in the Hg(II)/Hg(0) ratio in the plumes (Seigneur et al., 2001b).

Because Hg(II) reduction may be underestimated in current models, Hg wet deposition is likely to be overestimated. This means that the local and regional impacts of Hg emission sources are likely to be overestimated. Therefore, the simulations conducted in this study are likely to represent an upper limit on the Hg wet deposition values and of the contributions of local and regional sources of Hg deposition in Wisconsin.

# 4. CONCLUSION

TEAM was used to conduct a one-way nested grid simulation in which a fine grid with a horizontal resolution of 20 km was imbedded within the coarse 100 km resolution grid used in previous applications (Seigneur *et al*., 2002). The high-resolution fine grid covered Wisconsin, its neighbors and other states in the northeastern United States, and parts of Canada. The emissions inventory for the state of Wisconsin was modified to reflect the emissions inventory data available from the Wisconsin Department of Natural Resources. The simulation was conducted to study wet deposition in Wisconsin for the 1998 calendar year.

The spatial distributions of dry, wet, and total Hg deposition fluxes in Wisconsin were analyzed and a comparison made of model results with observations for the base case. Overall, model performance was considered satisfactory for wet deposition fluxes at the location of the four MDN stations in Wisconsin. A positive bias was exhibited in the model predictions of wet deposition, the possible reasons for which are discussed below. TEAM results were also compared with those from regulatory agency use of the EPA REMSAD and RELMAP models. TEAM showed better performance (where the performance criterion is a comparison between modeled and measured Hg annual wet deposition fluxes) than the other two models.

Three emission sensitivity simulations were also conducted over the fine grid. In the first sensitivity simulation, all anthropogenic Hg emissions from Wisconsin were set to zero. In the second sensitivity simulation, all anthropogenic Hg emissions from Wisconsin and all coal-fired power plant Hg emissions from Minnesota, Iowa, Illinois, Indiana, Michigan, Missouri, and Ohio were set to zero. In the third sensitivity simulation, all coal-fired power plant Hg emissions from Wisconsin were set to zero. The results have been analyzed in terms of changes in total annual Hg deposition between each sensitivity simulation and the base case.

Emissions from all anthropogenic sources in Wisconsin have an impact ranging from 4 to 10% at the Wisconsin MDN sites, less than 10% impact on total Hg deposition in northern Wisconsin, and less than 25% impact in most of central and southern Wisconsin. Coal-fired utility emissions in Wisconsin have an impact ranging from 1 to

4% at the Wisconsin MDN sites, and less than 5% impact on total Hg deposition in most parts of the state, with some isolated areas exhibiting 10 to 23% decreases in total deposition fluxes. Note, however, that the regional model used in this study is not suited to quantify local impacts and may overestimate such impacts. Emissions from utilities in some of the neighboring states of Minnesota, Iowa, Michigan, and Illinois have an impact ranging from 2 to 8% on Hg wet deposition at the Wisconsin MDN sites.

Results obtained from several independent experimental studies suggest that current models of the atmospheric fate and transport of Hg may underestimate the reduction of Hg(II) to Hg(0) in power plant plumes. Reduction of Hg(II) would lead to less deposition of Hg downwind of power plants since Hg(II) deposition dominates total Hg deposition near Hg sources. Therefore, the results presented here should be seen as upper limits of the contributions of local and regional sources to Hg deposition in Wisconsin.

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